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Doctoral Degree in Technology, Innovation, and Management

- XXXV Cycle -

DEVELOPMENT OF INNOVATIVE TEXTILE ARCHITECTURES BASED ON RECYCLED CARBON FIBERS FOR THE COMPOSITE MATERIAL INDUSTRY

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"However difficult life may seem,
there is always something you can
do and succeed at"

(Stephen Hawking)

"Success is the ability to go from
one failure to another with no loss
of enthusiasm"

(Winston Churchill)

"It always seems impossible until it's done."

(Nelson Mandela)

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In the blink of an eye this journey came to an end. It has been an intense period, made up of great satisfaction and happy moments, but also very difficult periods. Many times, I had the feeling of being alone. However, since I have always been used to fending for myself, I have tried every time not to drown and to do my best to resurface. I am deeply aware that it has been a troubled journey due to several external variables beyond my control. Nevertheless, I never gave up and I successfully achieved my goal. Obviously, I also recognise that each person I have met has contributed somehow to my growth. Therefore, it is now worth taking time to express my gratitude.

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ABSTRACT

The increasing use of carbon fibre-reinforced plastic composite materials in several industrial applications concerning, for instance, transportation, construction and luxury sport equipment has led to an increase of waste generated, which is mainly incinerated or landfilled, causing significant environmental issues. The main reason lies in the inability to effectively utilise recycled carbon fibres that currently may only be used for the production of second-quality materials such as non-woven fabrics and injection moulded composites which possess low mechanical properties. As yarns are characterised by high fibre orientation and good compactness, they can be exploited to manufacture more oriented reinforcements, potentially enhancing the performance of final composite materials. Therefore, spinning could represent a way to expand the use of recycled carbon fibre to more structural components. In such a context, this Ph.D. project aims at developing an innovative spinning process to obtain yarns with repeatable physical, thermal and mechanical properties suitable for the production of carbon fibre-reinforced plastic composites for structural applications. To address this goal, a six-stage approach was followed. The achieved results show several pieces of evidence. First, recycled carbon fibres from manufacturing scraps may be handled by the proposed innovative spinning process, but they require to be blended with a thermoplastic fibre. Second, manufactured hybrid yarns have good tensile properties and good amount of recycled carbon fibre, although the different steps composing the proposed innovative spinning process reduce the theoretical amount inserted; therefore, they could be adopted for the fabrication of good-quality composite materials. CFRPs consisting of hybrid yarns produced with 70% recycled carbon fibres appear to be the most promising from a mechanical perspective. Third, the sustainability of the innovative spinning process has been analysed. Eventually, it is worth noting that this thesis has both theoretical and practical implications. On the one side, it improves the knowledge about the relationship between composite materials and circular economy by expanding the knowledge about spinning of carbon fibres waste from manufacturing scraps and providing future research directions in the field of fibre-reinforced plastic composite materials recycling technologies. On the other side, it offers a strong practical enrichment to the composite material industry by bringing important economic savings to companies, which may reduce both their raw materials purchase costs and waste disposal costs. Furthermore, the

outcomes lead to a significant reduction in environmental impact, since the recycling of carbon fibres from manufacturing scraps allows the reduction of the use of virgin raw material and pollution due to incineration and landfill, as well as the recovery of the energy embodied in carbon fibres during their production.

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LIST OF ABBREVIATIONS

CF: Carbon fibre

CFs: Carbon fibres

CFRP: Carbon fibre-reinforced plastic

CFRPs: Carbon fibre-reinforced plastics

DSC: Differential scanning calorimetry

FRP: Fibre-reinforced plastic

FRPs: Fibre-reinforced plastics

GFs: Glass fibres

GFRP: Glass fibre-reinforced plastic

GFRPs: glass fibre-reinforced plastics

LCA: Life cycle assessment

LCC: Life cycle costing

eLCC: Environmental life cycle costing

TGA: Thermogravimetric analysis

CHAPTER 1

Introduction

1.1 Background and Motivation

Over the last decade, the concept of circular economy has been gaining momentum in academia, industry, as well as policymakers agenda (Burger et al., 2019). In fact, circular economy is the main way to encourage sustainable development (Ghisellini et al., 2016) which was defined by the Brundtland Commission in 1987 as a development path that meets the needs of the present without compromising the ability of future generations to meet their own needs (Geissdoerfer et al., 2017). However, a universal definition of circular economy has not yet been formulated to date. Although the term was originated in 1990 by D.W. Pearce and R. K. Turner, the most famous definition has been minted by Ellen MacArthur Foundation who states that circular economy is “an industrial system restorative and regenerative by intention and design” whose objective is to uphold products, parts and materials at their highest utility for as long as possible (Ellen MacArthur Foundation, 2013). Through closed-loop cycles of reuse, remanufacturing and recycling, circular economy decouples economic growth from resource consumption and environmental losses (Ghisellini et al., 2016). Therefore, it constitutes a viable alternative to the traditional linear production model, so called “take-make-dispose”, where products, obtained from virgin resources, are used and then disposed of in landfills after their use (Ellen MacArthur Foundation, 2013).

Nevertheless, this new economic paradigm is still far from being fully understood and applied widely. The main motivation underpinning this situation lies in the difficulties of managing this approach. Specifically, it is really challenging for a company to use as input something that is recognised by law as waste, but also to be aware of the actual availability of end-of-life waste or manufacturing scraps, since there is no dedicated supply chain to the collect materials for re-use, recovery or recycling (Yang et al., 2012). Moreover, when recycling is the only way forward, it is in many cases difficult to implement due to lack of technical capabilities. These challenges are particularly evident when dealing with composite materials (Mativenga et al., 2017).

Indeed, a composite material consists of two or more distinct materials with significantly different physical and chemical properties, whose combination enables the production of a new material with improved characteristics substantially different from those of the individual components (Hasan et al., 2019). Typically, such materials are composed of two constituents: a matrix, namely the continuous phase, and a reinforcement, namely the discontinuous phase. The former transfers the load to and between the reinforcements and protects them from the environment and handling. The latter, instead, provides strength, rigidity and hardness (Campbell, 2010; Haghshenas, 2016).

Composite materials may be classified according to the type of matrix or the type of reinforcement. The former can be a ceramic, a metal, or a polymer, while the latter can be in the form of fibres (short or continuous) or particulates (Figure 1). Among the others, polymer is the most widely used type of matrix (Sauer et al., 2018). In detail, thermosetting polymer represents the wide majority (about 80%) of the polymer matrix composite materials market (Mishnaevsky et al., 2017). Within this category, carbon fibre-reinforced plastic (CFRP) and glass fibre-reinforced plastic (GFRP) composite materials stand out. These materials are increasingly adopted in several industries, such as aircraft, automotive, and luxury sports equipment, owing to their outstanding

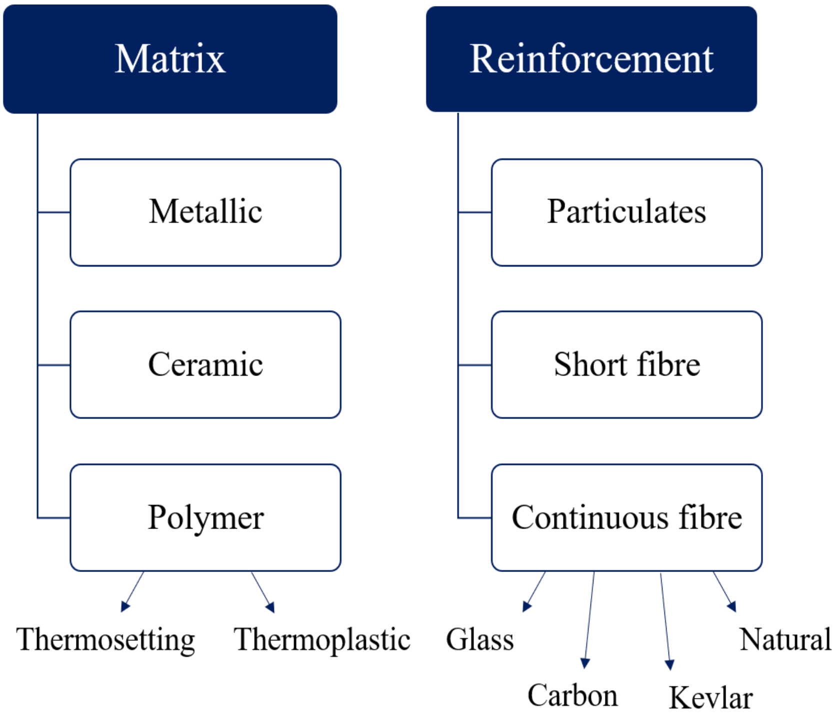

Figure 1: Types of matrices and reinforcements

properties, in terms of high mechanical strength and specific stiffness, and low density (Gemi et al., 2021; Zhang et al., 2020). These peculiarities allow companies to comply with increasingly stringent environmental constraints on CO_2 emissions. In 2017, approximately 1.12 million tons of GFRPs were produced in Europe, reaching the sixth consecutive year of growth. In the same year, the volume of global demand for CFRP was around 114 thousand tons, about 14% higher than the value reached the previous year (Sauer et al., 2018). Moreover, within the period 2010-2018, the total demand for CFRP has been characterised by a continuous expansion, with an average annual growth rate (CAGR) of 11.69% (Sauer, 2019), as shown in Figure 2.

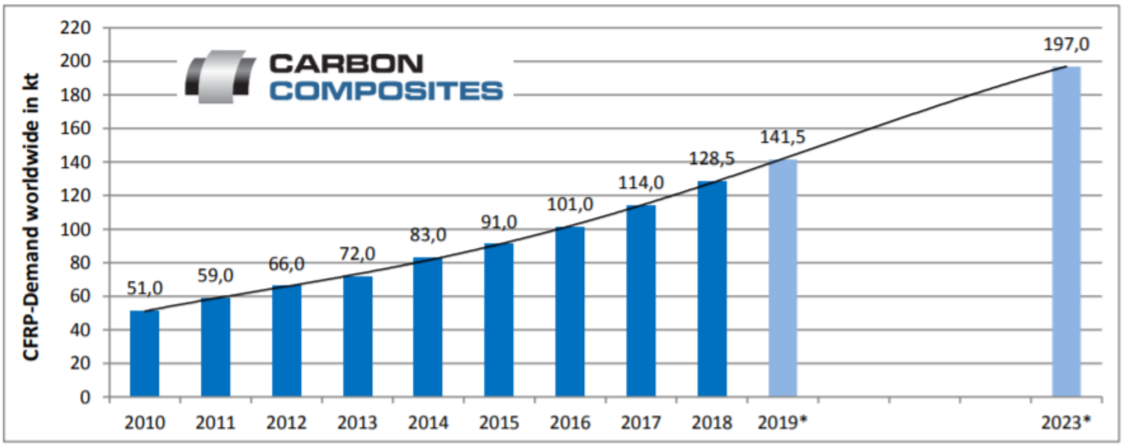

Figure 2: Global demand trend for carbon fibre-reinforced plastic matrix composite materials
Source: Sauer et al. (2019)

The massive use of fibre-reinforced plastic (FRP) composite materials has given rise to a huge increase in generated waste at different stages of their life cycle. In detail, around 40% of CFs is lost during the production phase (McConnell, 2010; Pimenta and Pinho, 2011), while losses of GFs generated during the production of GFRPs range between 5 and 10% (Bashir et al., 2017). Furthermore, as far as the end-of-life waste is concerned, it is expected that about 6000–8000 commercial airplanes will be decommissioned by 2030 (McConnell, 2010). All this waste has to be managed in order to prevent environmental issues (Lin and Schlarb, 2019), reduce economic losses for companies and provide added value to the overall society in the light of circular economy principles.

Although GFRPs account for more than 90% of all FRP composite materials produced (Sauer, 2019), the experimental part of this Ph.D. dissertation focuses on CFRPs. Indeed, they are composed of carbon fibres (CFs) which are a really valuable raw material from both a technical and economic point of view. More details on CFs are reported in the subsequent Section.

1.1.1 Carbon fibre

CF is defined as a fibre that contains at least 92% carbon by weight (Fitzer et al., 1989). However, when it encompasses at least 99% carbon, it is typically named graphite fibre. Owing to its chemical structure, CF presents an anisotropic behaviour (Huang, 2009), namely its characteristics depend on the direction along which they are considered. Other physical properties of CF include a diameter ranging between 5-15 μm and low density in the range of 1.75–2.00 g/cm^3 (Frank et al., 2012; Huang, 2009). Overall, CF possesses outstanding properties. First, it is characterised by a high strength to weight ratio; key aspect in improving CFRPs features (Bhatt and Goe, 2017; Pakdel et al., 2020). Second, it is rigid and brittle; therefore, breaks at a very low strain when bending. Third, it is characterised by good fatigue resistance and good tensile strength (Bhatt and Goe, 2017; Tehrani et al., 2013). Furthermore, CF is chemically stable and both abrasion and corrosion resistant, but also electrically conductive, and has low coefficient of thermal expansion (Bhatt and Goe, 2017; Clark et al., 2020; Frank et al., 2012).

The mechanical properties of CF ride on the kind of precursor used in its production (Pakdel et al., 2020). There are two types of precursors: polyacrylonitrile (PAN) and coal pitch. Specifically, PAN-based CF dominates the market with a share of 90% on the whole production (Bhatt and Goe, 2017; Frank et al., 2012; Huang, 2009). Their production process is similar, although different precursors require diverse processing conditions (Frank et al., 2012; Huang, 2009), and is composed of three main phases, namely: 1) Stabilization and Oxidation, 2) Carbonization, and 3) Graphitization. Initially, precursor fibres are stabilized in air through an oxidization process at around 200-400°C for 30-120 minutes to give the cyclic polymer structure. Then, during the carbonization phase, stabilized fibres are heated to a temperature of 1,000-1,700°C in an inert atmosphere to obtain clean and high-strength fibres. Sometimes, carbonized fibres are graphitized at temperatures of up to 3,000°C, in order to improve the mechanical characteristics of CF. In particular, by doing so, carbon content, as well as Young's modulus increase in the direction of the fibre. Finally, fibres are coated to improve the bonding phase with the composite matrices. This stage is named sizing.

Despite all the advantages, CF is a really expensive reinforcement (estimated prices range between €17-50/kg) owing to two main reasons. On the one hand, its production process is energy-intensive due to the oxidation of the precursor (Morin et al., 2012), requiring around 198-595 MJ/kg (F. Meng et al., 2018), and, therefore, possesses a high embodied energy that may not be wasted. On the other hand, the initial precursor is also very expensive. Indeed, it is responsible of about 50% of the cost of a conventional CF (Mainka et al., 2015).

1.2 Problem statement

Considering the huge amount of CF manufacturing scraps and CF end-of-life waste, to close the FRPs loop once and for all, many researchers are striving to find out ways to reuse them in structural applications. Specifically, on one hand, they are trying to improve the existing recycling technologies as well as to develop new and innovative ones in order to recover better quality fibres. On the other hand, they are committed in identifying and developing remanufacturing processes able to process both recycled CFs from end-of-life waste and from manufacturing scraps, which are naturally short and discontinuous, for the manufacture of value-added products. Currently, recycled CFs may be mainly used for the production of second-quality materials, including short fibres random mats (Hengstermann et al., 2017) and injection moulded composites (Hasan et al., 2018), that are not suitable for the production of structural applications.

According to numerous scientific studies (Akonda et al., 2012; Hasan et al., 2018; Hengstermann et al., 2016b), the spinning process might be a feasible solution to widen the application of recycled CFs to more structural components as it can be applied to short and discontinuous fibres to approach the properties of virgin tow. Indeed, the performance of CFRPs from recycled CFs could be drastically enhanced by adopting a reinforcement made up of yarns, which are the final outputs of the above-mentioned process, or their semi-finished products, since they are characterised by high fibre orientation and good compactness (Hengstermann et al., 2021). Nevertheless, regardless of their origin, recycled CFs do not possess the technical characteristics suitable for a traditional spinning process, as they are brittle, sensitive to shear stress, and without natural crimp (Hengstermann et al., 2016b). Therefore, the conventional spinning process is both hard and ineffective.

In such a context, the research and the development of innovative spinning processes for recycled CFs is vital to manufacture yarns with repeatable thermal-physical and mechanical properties (Hengstermann et al., 2021) and suitable for the reinforcement of CFRPs for structural applications, thus making value added use of waste and contributing definitively to closing the loop (Hengstermann et al., 2021; Stelzer et al., 2022). At this point, it is worth mentioning that, within this Ph.D. project, the focus was on the use of CF from manufacturing scraps for two main reasons. First, this type of source of waste accounts for approximately 40% of the total CFRPs waste produced (Pimenta and Pinho, 2011). Second, in the light of circular economy principles, it is good practice to implement reuse practices first and move to recycling only as a very last step (Ellen MacArthur Foundation, 2013).

1.3 Final aim and research questions

This research aims at developing an innovative spinning process able to produce a yarn made of recycled CFs with repeatable thermal-physical and mechanical properties and suitable for the production of CFRP composite materials for structural applications. Therefore, the focus of this Ph.D. project is on the topic of remanufacturing to provide value-added solutions (i.e. unidirectional thermosetting CFRPs) from a circular economy perspective. To this end, the path has been based upon four main research questions:

RQ1*: Which type of CF waste source has the best properties?*

RQ1.1: How have recycling technologies developed over time?

RQ1.2: What is the recycling technology with the greatest potential for growth?

RQ2: *How can the traditional spinning process be adapted to deal with recycled CFs?*

RQ3*: What are the characteristics of the potential CFRPs produced by the innovative process?*

***RQ4**: What is the environmental impact of the developed spinning process?*

The first research question arises from the presence of different potential sources of CF waste, namely manufacturing scraps, out-of-date prepregs and end-of-life CFRPs (Hasan et al., 2018; Khurshid et al., 2020). Each source of CF waste has distinctive characteristics and requires specific treatment methods for fibre recovering. In particular, CFs from manufacturing scraps, including offcuts, bobbin ends and selvedge, are resin-free and still sized materials and, accordingly, possess properties similar to the virgin CFs (Hasan et al., 2018). Therefore, no specific actions are compulsory to recover them (Hengstermann et al., 2021). At most, a mechanical opening operation could also be required to open up the orthogonal fabrics. Instead, CFs from out-of-date prepregs and end-of-life CFRPs are impregnated with resin and thus they have to be recovered through specific recycling processes (i.e. mechanical, thermal or chemical), leading to low property material. As a result, it is essential to study each recycling technology in depth.

Accordingly, Chapter 2 first provides an overview of the three existing recycling technologies. Then, it is divided into two main sections to address the sub-questions that arose. The first part is a bibliometric analysis using a systematic approach to answer RQ1.1, while the second part is a patent analysis to reply to RQ1.2.

So far, recovered fibres have mainly been used for the production of second-quality materials, including short fibres random mats (Hengstermann et al., 2017) and injection moulded composites (Hasan et al., 2018). Therefore, an innovative spinning process is required to produce CFRPs for structural applications; thus, expanding business opportunities and reducing incineration and disposal of high quantity of waste.

After developing the innovative spinning process and validating its technical feasibility, attention is turned to the production of CFRPs suitable for structural applications. To determine whether this goal may be accomplished, specific well-known laboratory tests are carried out.

At this point, it may be stated that the innovative spinning process is able to produce hybrid yarns suitable for the manufacture of CFRPs which can be applied in structural components. However, there is still an open point. It is not known whether the innovative spinning process developed is actually environmentally sustainable. Therefore, a specific environmental analysis is carried out.

1.4 Research design

The research questions have been addressed by leveraging on different rigorous methodologies. These have been chosen considering the technical/practical nature of this project. Indeed, this three-year period was jointly funded by Lombardy Region and ENEA (Italian National Agency for New Technologies, Energy, and Sustainable Economic Development) to address actual problems highlighted by composite material industry and, at the same time, reduce the environmental impact.

Especially at the beginning of the Ph.D., some methodologies related to the theoretical and academic world have been adopted to define the general purpose of the study. The first step consisted of a literature review aimed at identifying the main extant gaps on the topic at hand. More in detail, a bibliometric analysis using a systematic approach (i.e. PRISMA Statement (Moher et al., 2009)) was first carried out on papers about FRPs recycling technologies, as it allows investigating the advancement of science for a specific issue (Borri et al., 2021). In particular, it permits to conduct both quantitative and qualitative examinations to find out current hotspots and gaps in research and, accordingly, future avenues. Second, a patent-based analysis was performed using a patent technology roadmap (Mehrotra et al., 2016) to enhance knowledge about prior, current and future use of the main recycling technologies and, at the same time, provide a complementary perspective to the results obtained from the previous investigation. Both methodologies are described thoroughly in Chapter 2. Since it was found that scant attention has been paid to the production methods of FRP composites using recycled fibres for structural applications, in the subsequent steps, the work focused on more experimental activities.

Specifically, to address the specific need that emerged in the first step of the Ph.D. project, the researcher focused on the topic of technology innovation. In greater detail, the traditional spinning process was innovated by following a reasoned ‘trial-and-error’ approach. Likewise, to assess their quality, the final hybrid yarns and the unidirectional thermosetting CFRPs were tested at laboratory level leveraging standard established methods. Concurrently, an experimental campaign on the base of an unbalanced full factorial design of experiments (DoE) considering specific parameters, such as the amount of recycled CFs, the type of thermoplastic fibre and the number of draw frame doubling on two or three levels was designed to statistically analysed the obtained results. Specifically, the analysis of variance (ANOVA) was adopted to verify whether these affect the final amount of recycled CF present in the hybrid yarns as well as the tensile properties of the final CFRPs.

Lastly, a Life Cycle Assessment (LCA) analysis has been carried out by following the specific and well-recognised ISO 14040 standard to evaluate the actual sustainability level of the proposed innovative spinning process. In detail, a cradle-to-gate approach has been selected and modelled using the attributional LCA.

1.5 Research outline

Figure 3 sketches the research structure in terms of research questions, specifying the chapters in which each of them is discussed.

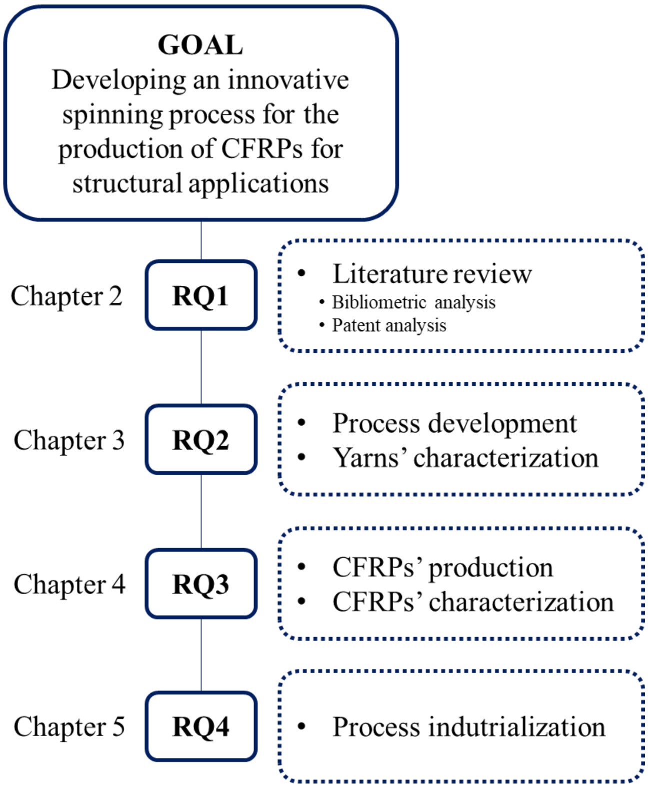

Figure 3: Research breakdown structure

Overall, the thesis consists of six chapters. After this introduction (Chapter 1), the dissertation is structured as follows. Chapter 2 addresses the first research question by providing a thorough analysis of the existing recycling technologies. It is divided into two main sub-sections. The first one concerns a bibliometric analysis whose papers were gathered using a systematic approach and the second one a patent trend analysis. Chapter 3, instead, explains the developed innovative spinning process and proposes the characterisation of the hybrid yarns produced at laboratory level. While Chapter 4 reports on both the manufacture and characterisation of the CFRPs obtained. Furthermore, Chapter 5 presents the LCA performed to evaluate the environmental sustainability of the innovative spinning process. Finally, Chapter 6 ends the thesis by drawing the main conclusions, discussing limitations and potential future improvements.

1.6 Research contribution

1.6.1 Academic contribution

This thesis contributes to the existing scientific literature in different ways. First, wider knowledge about the spinning of CFs waste was developed in this Ph.D. project. Indeed, an innovative spinning process was proposed and its validity was assessed by a technical feasibility study which involved different types of thermoplastic fibres and blending ratios. Furthermore, more in-depth knowledge of its potential was gained thanks to the actual production and subsequent characterisation of unidirectional thermosetting CFRPs. In doing so, this thesis appears to be one of the first attempts to answer the gap that emerged from the literature review concerning the need to study the feasibility of manufacturing methods that adopt recycled fibres (Colombo et al., 2022). Moreover, it strongly contributes to the literature on technological innovation in textile industry as well as the sustainability of composite materials. As regards the latter, indeed, the outcomes of this project prove the possibility to apply circular economy principles to this industry.

Furthermore, to the best of researcher's knowledge, this thesis provides the first study on the environmental sustainability of processes that can process recycled CF to obtain semi-finished products with good specific properties that in turn can be used for value-added applications. In this way, a first step was taken towards the concept of eco-efficiency within the composite materials industry.

Finally, another major implication concerns the enrichment of knowledge about recycling technologies for the recovery of CFs and GFs. In detail, by means of the patent analysis and the bibliometric analysis, a complete and critical overview of the existing recycling technologies and an assessment of the actual maturity level of each of them was provided. Moreover, hotspots and gaps, as well as future research directions were identified.

1.6.2 Industrial contribution

This thesis offers a strong practical enrichment to all the companies belonging to the composite material industry from both a financial and a strategical point of view. By allowing the use of recycled CFs in light of circular economy, it brings important economic savings for composite manufacturers. Indeed, they should no longer incur landfill costs and, at the same time, could benefit from the sale of manufacturing waste to those who recycle it to recover CFs and manufacturing scraps to those who reuse them directly. Furthermore, the possibility of producing CFRPs using reinforcement composed of recycled initial fibres aligns them with the principles of sustainability, thus increasing their visibility.

The same evidences might be applied to other upstream actors in the supply chain. For instance, the companies producing the yarns that are used to fabricate the reinforcements and the companies producing the reinforcements themselves could reduce their raw materials purchase costs, since recycled CF has lower purchase prices than virgin CF (as an example, estimated prices of virgin CF range between €17-50/kg, while the cost of recycle CF is around €8-12/kg) (Fernández et al., 2021; van de Werken et al., 2019). In doing so, these companies contribute in a more or less conscious way to the topic of circular economy. CF producers, instead, would have less of an environmental impact as the demand for virgin CF could decrease globally. This situation could represent an opportunity for these companies to convert part of their business model into technologies for recycling end-of-life CFRPs or manufacturing waste. In doing so, they could reduce their energy costs, as recycling CF requires less energy than producing virgin fibre.

Finally, taking a broader perspective, the possibility of using recycled CFs from manufacturing scraps for the production of CFRPs potentially decreases the environmental impact due to the production of virgin CF, since the carbonisation phase, which is responsible for more than 50% of the climate impact and energy use of CFRP production (Hermansson et al., 2019), is omitted. Besides, it reduces the impacts owing to incineration and landfilling of the composite material industry.

1.7 Research dissemination

During the three years of the Ph.D. program, different scientific contributions on the topic at hand have been submitted to or published on international peer-reviewed journals. It is worth mentioning that different parts of the present dissertation draw from these studies. They are listed in the following divided by the part of the manuscript to which they contributed.

Research question 1

- Colombo, B., Gaiardelli, P., Dotti, S., Caretto, F., Coletta, G., 2021. Recycling of Waste Fiber-Reinforced Plastic Composites: A Patent-Based Analysis. *Recycling* 2021, Vol. 6, Page 72 6, 72. https://doi.org/10.3390/RECYCLING6040072;
- Colombo, B., Gaiardelli, P., Dotti, S., Caretto, F., 2022. Recycling technologies for fibre-reinforced plastic composite materials : A bibliometric analysis using a systematic approach. *Journal of Composite Materials* 0, 1–18. https://doi.org/10.1177/00219983221109877

Research question 2

- Colombo, B., Abdoos, A., Gaiardelli, P., Dotti, S., Caretto, F., 2021. A technical feasibility study of an innovative spinning process for recycled carbon fibres. *26th Summer School Francesco Turco,* 2021. AIDI-Italian Association of Industrial Operations Professors 2021.
- Colombo, B., Gaiardelli, P., Dotti, S., Caretto, F., 2022. Analysis of the physical, chemical and mechanical properties of hybrid yarns for good-quality recycled carbon fibre-reinforced plastic composites. *27th Summer School Francesco Turco,* 2022. AIDI-Italian Association of Industrial Operations Professors 2022.
- Colombo, B., Gaiardelli, P., Dotti, S., Caretto, F., 2023. An innovative spinning process for production and characterisation of ring-spun hybrid yarns from recycled carbon fibre. *Journal of Cleaner Production,* 137069.

Research question 3

- Colombo, B., Gaiardelli, P., Dotti, S., Caretto, F. Tensile properties of plastic composites reinforced with hybrid yarns from recycled carbon fibers. (ongoing). Journal target: *The Journal of The Textile Institute*.

Research question 4

- Colombo, B., Gaiardelli, P. Environmental assessment of a spinning process for the production of ring-spun hybrid yarns from recycled carbon fiber: a cradle-to-gate approach. Journal target: *Journal of Cleaner Production*.

Moreover, research extra to the topic in question was published during the Ph.D. span. They are listed in the following.

- Colombo, B., Boffelli, A., Gaiardelli, P., Kalchschmidt, M., Madonna, A., Sangalli, T., 2022. A Multiple Case Study on Collaboration for a Circular Economy: A Focus on the Italian Textile Supply Chain. *IFIP Advanced Information Communication Technologies* 663 IFIP, 408-415.
- Colombo, B., Madonna, A., Boffelli, A., Gaiardelli, P., 2022. Supply Chain ed economia circolare: ESEMPI DAL SETTORE tessile-abbigliamento. *Logistica Management*. 324, 42-48.
- Trussardi, D., Lumina, A., Colombo, B., Boffelli, A., Gaiardelli, P., 2022. Progettazione sostenibile, un metodo innovativo per ridurre l’ impatto ambientale dei prodotti. *SISTEMI&IMPRESA* 3, 32–37.
- Colombo, B., Abdoos, A., Dotti, S., Gaiardelli, P., 2022. Textile Machinery Industry in the Era of Digital Transformation. Adv. Sci. Technol. 113, 113–120.
- Colombo, B., Gaiardelli, P., Dotti, S., Boffelli, A., 2021. Business Models in Circular Economy: A Systematic Literature Review. *IFIP Advanced Information Communication Technologies* 632 IFIP, 386–393.
- Colombo, B., Gaiardelli, P., Dotti, S., 2020. Overcoming barriers of green transformation through the adoption of lean manufacturing: A case study. *25th Summer School Francesco Turco*, 2020. AIDI-Italian Association of Industrial Operations Professors 2020.
- Colombo, B., Boffelli, A., Gaiardelli, P., Dotti, S. (2020). Esempi virtuosi dal settore tessile e abbigliamento. *Logistica Management*. 80-85

Lastly, it is worth mentioning that most of the studies presented in this dissertation were improved through the different feedbacks received from scholars experienced in the field under investigation thanks to participation in national and international conferences, as well as Ph.D. workshops.

International and national conferences

- APMS Conference 2022, Gyeongju, Korea, September 25-30, 2022;
- XXVII Summer School Francesco Turco, Riviera dei Fiori, Italy, September 7-9, 2022;
- Circular Economy in the Era of Digitalization, Brescia, Italy, June 10, 2022;
- 9th EurOMA Sustainability Forum, Zagreb, Croatia, March 21-22, 2022;
- XXVI Summer School Francesco Turco, Bergamo, Italy, September 08-10, 2021;
- APMS Conference 2021, Nantes, France, September 05-09, 2021;
- XXV Summer School Francesco Turco, Bergamo, Italy, September 09-11, 2020.

Ph.D. workshops

- APMS 2022 Marco Garetti Doctoral Workshop, September 25, 2022;
- IX PhD On the Go, Benevento, Italy, June 16-17, 2022;
- APMS 2021 Marco Garetti Doctoral Workshop, September 5, 2021;
- VIII PhD On the Go, Benevento, Italy, June 24-25, 2021.

CHAPTER 2

Literature review

2.1 Objectives

Since the starting point for developing the innovative spinning process is represented by the recycled CFs, the present chapter aims to perform an in-depth analysis of the existing recycling technologies by providing an overview of them in order to determine the properties of the CFs derived from each recycling technology, as well as to assess the actual maturity level of each of them. As an initial point, the study took a broader perspective in order not to exclude essential information. Specifically, contributions concerning CFs and GFs were considered, as both are worth studying due to different aspects. In detail, GFs are exploited to produce GFRPs which are widely utilized in final applications. Indeed, GFRP composites account for more than 90% of all FRP composites produced (Sauer, 2019). While CFs are expensive and possess high embodied energy owing to their production process (F. Meng et al., 2018).

To this end, a bibliometric analysis using a systematic approach and a patent analysis have been carried out. Overall, by leveraging on these two approaches, the researcher is able to provide a complete and critical overview of the existing knowledge on the topic at hand and identify literature hotspots and gaps, as well as future research directions. Besides the significant theoretical value, this part of the dissertation has been also fundamental in understanding where to focus the more practical research effort. Indeed, focusing on designing an innovative spinning process on the basis of recycled CFs obtained by means of a technology which has little potential for future growth would likely not guarantee a solution that maximises the potential of the recovered material.

The remainder of this Chapter is organised as follows. First, an overview of each existing recycling technology is provided. Then, the Chapter is divided into two main sub-sections. One is dedicated to the bibliometric analysis using a systematic approach and the other to the patent analysis. In both cases, the methodology used and the outcomes obtained are explained in detail.

2.2 Theoretical background

Mechanical, thermal and chemical are the three major macro-categories of FRP composite materials recycling technologies (Pickering, 2006). The adoption of a specific technology rides on the material that needs to be recycled as well as the application for which it will be reused (Oliveux et al., 2015). The following section provides a description of each macro-category.

2.2.1 Mechanical recycling

Crushing or shredding huge waste/scraps into smaller parts (50–100 mm) is the initial step in mechanical recycling of FRP composites materials. The pieces are then further ground (10 mm–50 mm) in a high-speed mill or a hammer mill. Lastly, recycled materials, such as powders and coarse particles, are sieved according to their size (Pickering, 2006). The usage of recycled powders is commercially inconvenient, especially when considering CFRPs, due to the low cost of virgin fillers. Nevertheless, because of their resin content, they can be used as energy sources (Oliveux et al., 2015). Owing to the difficulty of adhesion between the recycled product and the virgin polymer, reuse of fibrous fractions is problematic and results in a significant decrease in mechanical properties (Pickering, 2006). Notwithstanding, mechanical recycling is still regarded as the best choice for GFRPs recycling in the scientific literature (Oliveux et al., 2015; Ribeiro et al., 2015) due to its low cost. On the other hand, it is not thought to be worthwhile for CFRPs recycling because of the high value of CFs.

2.2.2 Thermal recycling

Thermal recycling techniques such as pyrolysis and fluidized bed allow for the recovery of fibres by breaking down the polymer matrix. These reactions, according to Oliveux et al. (2015), occur at temperatures ranging from 450°C to 700°C, depending on the type of resin. Resin is volatilized, resulting in not only gases but also oil fractions and, in rare situations, char on fibres. The presence of char on recycled fibres might diminish their mechanical qualities and hence reducing their potential reuse (Pickering, 2006).

2.2.2.1 Pyrolysis

Pyrolysis allows the degradation of a polymer matrix by heating the material in the near absence of oxygen. During the process, gases, oil and chars are produced. In detail, chars contaminate fibre surface, making recovery difficult. To burn it and obtain clean fibres and fillers, post-pyrolysis treatments, such as oxidation processes, are required (Oliveux et al., 2015). These treatments, however, worsen the quality of the fibres. Depending on the operating temperatures and the type of fibres, mechanical, electrical and surface properties of recovered product vary (L O Meyer et al., 2009). This means that operating temperatures and other process parameters have to be optimized considering the trade-off between an efficient fibre recovery and acceptable mechanical properties (Oliveux et al., 2015; Poulikakos et al., 2017). Overall, the higher the operating temperature, the cleaner the fibre surface, but the lower the mechanical properties As far as epoxy resin is concerned, literature suggests temperatures ranging from 500-600°C in oxidant environment to completely removed the matrix without compromising fibres' properties (Meyer et al., 2009; Oliveux et al., 2015). Reclaimed fibres are discontinuous, short and fluffy; however, they exhibit modulus and tensile properties similar to virgin fibres, particularly for CFs (Giorgini et al., 2015; Naqvi et al., 2018). As a result, they can be deployed as raw material in the production of novel composites if appropriately handled.

Pyrolysis is the most widely utilized thermal process in commercial applications as it represents the most reliable and efficient process as regard energy and fibre recovery (Witik et al., 2013). Moreover, it enables the reclamation of CFs with excellent mechanical properties (Asmatulu et al., 2014; Pimenta and Pinho, 2011). Unfortunately, it is not the same for GFs that degrade by at least 50% (Cunliffe and Williams, 2003) due to high temperatures. This is the main reason why pyrolysis is principally exploited to recycle CFRPs (Oliveux et al., 2015; Yang et al., 2012). Nonetheless, there are studies on the pyrolysis process for GFRPs recycling in the scientific literature (e.g. Naqvi et al. (2018); Pickering (2006)).

2.2.2.2 Fluidized bed

Fluidised bed is another thermal recycling process. It has been developed for the first time at the University of Nottingham by Pickering et al. (2000). FRP composite material scraps are fed into a bed of silica sand that is fluidized by a high-temperature stream of air. As a result, the process takes place in an oxidant environment. The air heats and decomposes the composite material's polymer matrix, allowing the fibres and fillers to be released and carried away by the stream. They are then separated from the gas stream in a cyclone.

Finally, to obtain clean fibres, they must be sent through a secondary combustion chamber with a temperature of around 1000°C. During the process is produced a clean fuel gas which can be recovered as energy source. Reclaimed fibres, instead, fluffy and short with a mean length of 6-10 mm (Pickering, 2006). As regards GFs, at 450°C their tensile strength is reduced by 50%, while stiffness is comparable to virgin GFs. In the case of CFs, instead, a loss of tensile strength of about 25% results after processing at 550°C, whereas stiffness remains unchanged and surface shows a little degradation in oxygen content (Hyde et al., 2006). Tolerance to mixed and contaminated materials is the greatest advantage of this process. Indeed, heavier metal materials sink into the sand bed and do not blend with fibres and fillers (Pickering, 2006). Currently, the fluidized bed is mainly used to recycle GFs. CFs are damaged to a greater extent than when they are recovered by pyrolysis (Oliveux et al., 2015).

2.2.3 Chemical recycling

Chemical recycling refers to any method that uses a chemical reagent to degrade a polymer matrix in order to recover its fibres. When employing water, glycols, or acid as solvents, the depolymerization process, also known as solvolysis, can be classified into three sub-categories, namely hydrolysis, glycolysis and acid digestion. To dissolve the resin correctly and achieve high levels of efficiency, each type of solvent requires specific temperatures and pressures (Dang et al., 2005). Nonetheless, since it functions at lower temperatures, solvolysis is naturally less energy intensive than pyrolysis, especially for epoxy resins and unsaturated polyester (Oliveux et al., 2015). Furthermore, using water and alcohol as solvents is environmentally friendly because these components are non-hazardous and can be recovered by evaporation and distillation after the reaction (Yang et al., 2012). Solvolysis enables not only the recovery of clean fibres and fillers, but also the recovery of monomers from resin depolymerisation (Yang et al., 2012). Recovered fibres have high mechanical properties and fibre length (Pimenta and Pinho, 2011), but they suffer from a reduction in bonding to the polymer matrix (Jiang et al., 2009). Finally, chemical recycling has minimal tolerance to contamination (Marsh, 2009; Yang et al., 2012).

2.2.4 Recycling technologies: Benefits and drawbacks

Scientific studies show that mechanical recycling is the greatest option for GFRPs, whereas pyrolysis and solvolysis are the best options for CFRPs, given the high economic value of CFs and the decline in GFs' mechanical qualities owing to thermo-chemical processes. Nonetheless, it is evident from the literature that the right technique depends on the material to be recycled and its reuse applications (Oliveux et al., 2015). Furthermore, each macro-category has its own set of advantages and limitations, as shown in Table 1.

Table 1: Advantages and drawbacks of recycling processes (Colombo et al., 2021)

	Advantages		Drawbacks	
Mechanical recycling	Relatively simple	(Pickering, 2006)	Decrease of mechanical properties	(Pickering, 2006; Ribeiro et al., 2015)
	Environmentally-friendly	(Pimenta and Pinho, 2011)	Recovery of both fibers and resin as powders and coarse	(Palmer et al., 2010; Pickering, 2006)
			Few applications for remanufacturing	(Pimenta and Pinho, 2011)
Thermal recycling	High mechanical properties	(Giorgini et al., 2015; Pimenta and Pinho, 2011)	Fiber quality depends on process parameters	(L O Meyer et al., 2009; Poulikakos et al., 2017)
	Environmentally-friendly	(Pimenta and Pinho, 2011)	Recycled fibers in fluffy form	(Pickering, 2006)
	With pyrolysis, production of oil from the polymer matrix	(Pickering, 2006)	With pyrolysis, possibility of char on fiber surface	(L O Meyer et al., 2009)
	With fluidized bed, clean fiber surface and good tolerance to contaminated materials	(Pickering, 2006)	With fluidized bed, no recovery of polymer matrix and more decrease of fibers strength and length	(Oliveux et al., 2015; Pickering et al., 2000)
Chemical recycling	Recovery of both fibers and resin (the latter as monomers)	(Yang et al., 2012)	Generally, not environmentally-friendly	(Liu et al., 2004; Oliveux et al., 2015)
	High mechanical properties and fiber length	(Pimenta and Pinho, 2011)	Low tolerance to contaminated materials	(Yang et al., 2012)
			Reduction in bonding between fibers and new resin	(Jiang et al., 2009)

A recent study has also proposed that a new hybrid technique would be the best way to solve the drawbacks of thermal and chemical procedures. For example, Adherent

Technologies, a company of the United States, demonstrated that a three-step method consisting of a thermal pre-treatment followed by two solvolysis processes, the first at a low temperature and the second at a high temperature, may generate high-quality recycled fibres, particularly for CFs (Gosau et al., 2006).

2.3 Bibliometric analysis

2.3.1 Methodology

To find out hotspots, current and upcoming research trends, existing research gaps, as well as to understand the mechanical and chemical-physical properties of the CFs from each recycling technology a bibliometric analysis using a systematic approach has been carried out. Indeed, it has been identified as the best alternative for pursuing the research objective, since it is a technique that allows researchers to evaluate the advancement of knowledge for a specific subject (Borri et al., 2021). In detail, bibliometric analysis allows researchers to perform both quantitative and qualitative assessments to find existing research hotspots and gaps, as well as future potential directions. VOSviewer (version 1.6.17) was used to achieve this goal. VOSviewer is an open-source program that may be adopted to design, visualize and analyse bibliometric networks that consider the relationships between various pieces of scientific research. Simultaneously, a systematic approach to material collection was carried out to assure a repeatable and reliable process. It also enables authors to get over the limits of narrative reviews (Tranfield et al., 2003). In particular, the material collection was organized according to the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) statement, proposed by Moher et al. (2009). In order to ensure transparency and clarity, the PRISMA standards prescribe a precise set of measures to be followed. The four main phases are: (1) identification, (2) screening, (3) eligibility and (4) inclusion, have been addressed (Figure 4).

In summary, the whole procedure is composed of four main steps. The execution of the systematic data collection method is the first phase. The second step consisted in putting in practice the descriptive statistics of the bibliometric analysis into practice (i.e. performance analysis). The publication trend, journal allocation, published articles per year for the major countries and the most commonly used research types were all examined (Cobo et al., 2011; Noyons et al., 1999). The execution of the science mapping of the bibliometric analysis is the third phase. In detail, a keyword co-occurrence analysis and a document co-citation analysis were performed. Lastly, the final phase entails a discussion of the results presented by VOSviewer to highlight the most important research areas and suggest future research avenues after analysing the clusters suggested by the software (Rialti et al., 2019).

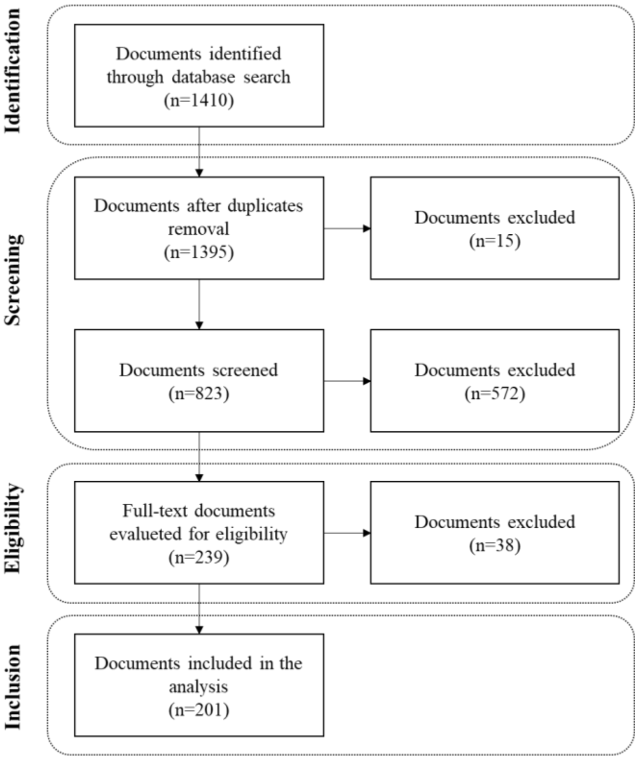

Figure 4: The review process in accordance with the PRISMA guidelines

2.3.1.1. Data sample

Scopus was used to gather the data as it is widely regarded as the most comprehensive and detailed scientific database (Chadegani et al., 2013). Indeed, Scopus is distinguished by a high level of uniqueness (Sánchez et al., 2017) as well as a broad data coverage (Salim et al., 2019).

The following query was set up in order to identify a sample batch of documents for investigation:

TITLE-ABS-KEY ((("fiber-reinforced plastic*" OR "fibre-reinforced plastic*" OR "fiber-reinforced polymer*" OR "fibre-reinforced polymer*" OR "thermoset composite material*" OR "thermoplastic composite material*") AND ("recycle" OR "recycling" OR "recover"

OR "recovering" OR "reclaiming" OR "reclamation"))) AND (EXCLUDE (PUBYEAR , 2022)) AND (LIMIT-TO (LANGUAGE , "English")) AND (LIMIT-TO (SRCTYPE , "j")) AND (LIMIT-TO (DOCTYPE , "ar") OR LIMIT-TO (DOCTYPE , "re"))

“Title, Author keywords, Abstract” were included in the search record. On September 17, 2021, the first round was conducted without any selection criteria and yielded 1410 outcomes. After removing duplicates, there were 1395 items left. The search was then limited to the English language, as it is the international academic language (Genç and Bada, 2010) and the year of publication was set to 2021. Furthermore, the research was confined to studies published in journals in order to maintain control over the quality of the publications (Light and Pillemer, 1984). This resulted in the examination of 823 papers. Articles were evaluated in three rounds, progressively increasing the content level under inspection, in addition to the filters already stated. First, titles were read, followed by abstract reading, which helped to narrow the pool even more, and last, whole texts were evaluated. Articles that did not suit the research's goal were excluded after title and abstract screening (584 articles). Then two qualifying criteria were established: only research relating to the explanation or enhancement of one or more recycling processes for FRP composite materials analysis was considered. This final stage resulted in the removal of 38 items. For the analysis, a total of 201 publications were considered.

Following that, data mining from the selected papers was accomplished using a data extraction form that served as a repository for eventual data synthesis. To keep inter-coder agreement in the 85-90 % range, a colleague of mine and I coded all of the articles and another author assessed the coding. Moreover, articles marked by dispute or uncertainty were discussed among us and agreement reached by consensus in order to ensure a high level of inter-rate dependability.

2.3.1.2 Descriptive statistics and science mapping of the bibliometric analysis

As regards the performance analysis, to enable descriptive and methodological assessments of the field, four aspects were defined and organized into two unique groups to provide a sound overall understanding of the topic. To define the characteristics of articles and assess the main trends in the literature, the time distribution of publications, paper origins and journal names, while using selected research methodologies to establish the methodological assumptions made were analysed. Instead, as regards science mapping, keyword co-occurrence analysis and document co-citation analysis were carried out.

The study of keywords is critical in advancing scientific research. Keywords assist scientists correctly study the evolution of a certain research stream (Barki et al., 1993) by specifying significant concepts in the topic at hand. Because of this, a co-occurrence analysis was used to look into the correlations between the keywords found. The research was conducted using the VoSviewer software (version 1.6.17) and it was based on Scopus’ "authors' keywords" field using the full counting approach. The analysis was limited to 180 publications since papers without keywords (i.e. 21 articles) were omitted. Furthermore, since authors often use slightly different terms to communicate similar or identical concepts, selected keywords were manually filtered for similarity (for instance, "carbon fiber" versus "carbon fiber" or "electrodynamic fragmentation" versus "high-voltage fragmentation") (Eck and Waltman, 2014). To construct a clear and complete network map, a minimum number of occurrences of a keyword equal to 6 was set up. There were 19 keywords that met the threshold. In addition, the classification proposed by Fadlalla and Amani (2015) was used to investigate the topic's evolution further. Persistency and incidence factors were considered in this regard. The former represents the intensity with which a keyword is used throughout time, i.e. the number of years a keyword has been used, whereas the latter, calculated as the ratio between the total number of times a keyword has appeared and its persistency, indicates the number of times a keyword has occurred. As a result of this normalization, the information contained in the incidence index can be decoupled from the information provided by persistence. Since the maximum persistency of the keywords in the examined documents was 19, the threshold for high and low persistency values was set at 9.5. In terms of incidence, the median value of 1.8 was used as the cut-off point to divide keywords in half and put them into high and low incidence categories.

A co-citation analysis was then utilized to investigate the relationship between different articles, propose a classification of studies on FRP recycling technologies, and comprehend the most important publications in the past. Co-citation was first defined by Henry Small in 1973 (Small, 1973) as the occurrence of two articles being cited together by another document: the higher the co-citation strength of two articles, the more likely they are semantically interrelated (Small, 1973), and thus belong to the same field of research (Fang et al., 2018; Hjørland, 2013). A thesaurus file was constructed to standardize citations written in various styles (Eck and Waltman, 2014). The investigation was then carried out using the VoSviewer software (version 1.6.17). The minimum number of citations required for a cited reference was set to 25. Only 18 papers were accepted. The VOSviewer output is a network with 18 nodes and 153 linkages, each of which is identifiable by the initial author's name and the year of publication. The thickness of links between documents shows the relative frequency with which the two documents were co-cited in the papers in the database, while the size of the nodes represents the relative frequency of document co-citation. The close proximity of two articles in the map suggests that they are regularly co-cited by other writers, implying a deeper intellectual link (Trujillo and Long, 2018).

2.3.2 Outcomes and related discussion

2.3.2.1 Performance analysis

Over time, there has been a rise in scientific interest in FRP recycling technologies. Figure 5 shows that from 1995 to 2011, research on the topic under investigation produced no more than four publications per year, but since 2012, the number of papers published has increased, reaching a peak of 34 papers in 2020. Additionally, nearly 62% of the articles were published between 2016 and 2020. Such sustained attention in recent years demonstrates the importance of this new subject of study, which merits further exploration. This upward trend could be influenced by a variety of factors. To begin with, law has gradually pushed for environmental sustainability (Kumar et al., 2019). Moreover, society as a whole is becoming more conscious of environmental issues (Brochado et al., 2017).

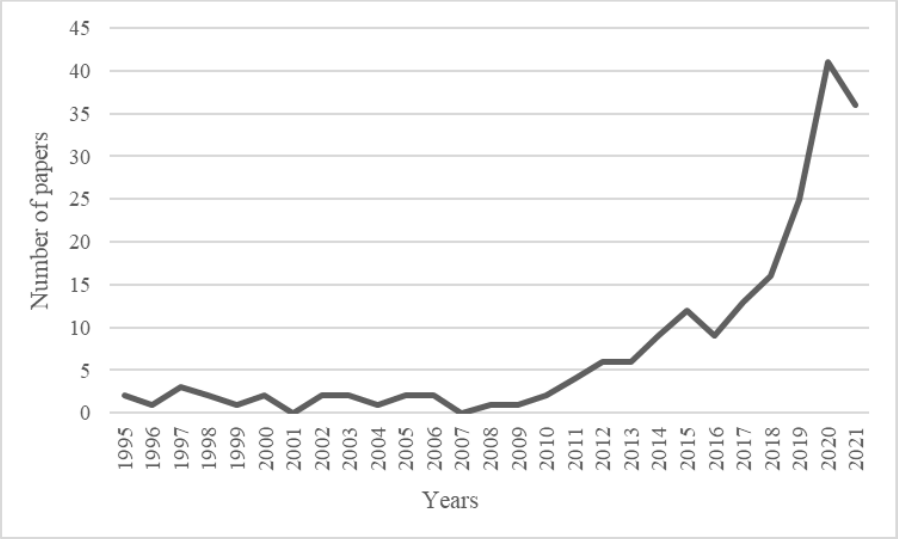

Figure 5: Papers' publication trend on FRP recycling technologies

Customers, whether end users or other businesses, are urged to own items that adhere to the concepts of sustainability and circular economy for a variety of reasons, including market competitiveness and social status (Ali et al., 2019; Miranda et al., 2019). Besides, businesses must develop technology solutions to improve production efficiency and, as a result, reduce expenses. Finally, the ability to recycle large quantities of fibres would allow businesses to be less reliant on market fluctuations. Indeed, the price of virgin raw materials is closely linked to the price of oil, which is renowned for its great volatility (Ellringmann et al., 2016).

Figure 6 shows the papers published per year on FRP composite materials recycling technologies for the major countries. It can be seen that papers about FRP began to be published in Europe before the 2000s, with a sharp increase especially after 2014, whereas research in the US and Japan started shortly after and in China only in 2009. The main driver of European interest in the topic may be traced back to the European Union's various waste management legislations, which aim to prevent or minimize environmental damage as much as possible. Moreover, the European Union plans to eliminate garbage disposal by 2025, necessitating the establishment of new and ground-breaking recycling pathways. Even in China, Japan and the United States, research was primarily motivated by environmental aims. However, it began later and was mostly based on a bottom-up strategy. In the United States, for example, such activities were mostly initiated by private enterprises due to the lack of particular legislation for end-of-life recycling (Miller et al., 2014). Overall, Europe is leading the research with the most documents available in the literature, but China is growing at the fastest rate. This latter conclusion is probably connected to the necessity to manage large amounts of waste owing to the high demand for FRP composite materials as a result of the country's rapid industrialization (Mordor Intelligence, 2020).

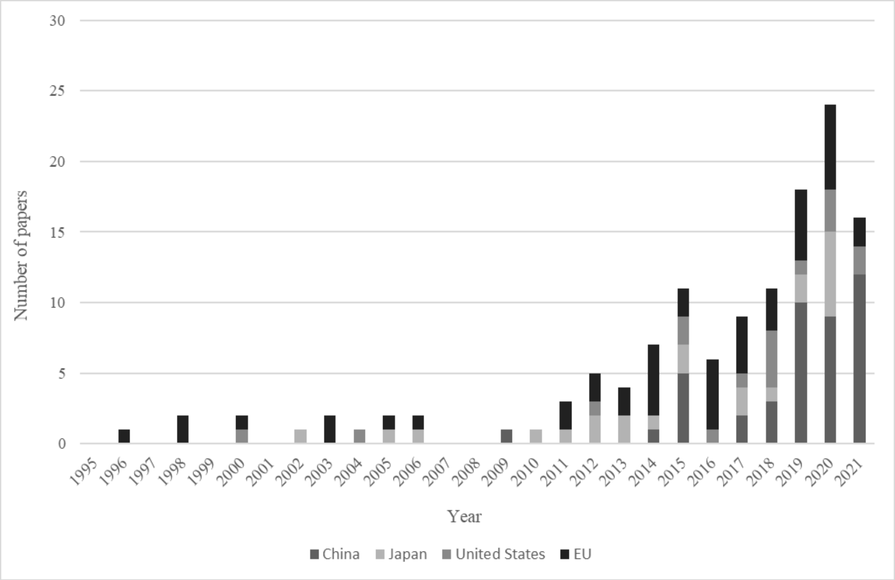

Figure 6: Papers' publication trend per country on FRP recycling technology

According to a descriptive investigation, journals publishing articles on the subject are extremely specific, and the majority of them have a technical imprint, as seen in Figure 7. The most common topics, for example, include chemistry-related problems, composite materials business and sustainability issues in various contexts. This information demonstrates that the subject under inquiry is growing increasingly interesting as the importance of environmental sustainability grows.

The articles were then evaluated based on the sort of research conducted (Figure 8). After being tailored to the context, the four analytical categories given by Merli et al. (2020), namely Review, LCA, Experimental investigation and Modelling, were used. Review studies are investigations whose major objective is to summarize previous research. LCA studies are studies that use a life cycle assessment tool to investigate the environmental impact of FRP recycling technology. Experimental studies are works that use experiments and tests to examine the mechanical, physical and chemical qualities of recycled fibres. Finally, modelling studies provide statistical/mathematical models to simulate the disintegration of the plastic matrix or evaluate the energy demand of recycling technology. In addition, a fifth category, namely, Theoretical, was created to include research that have no empirical contribution but are more theoretical in essence.

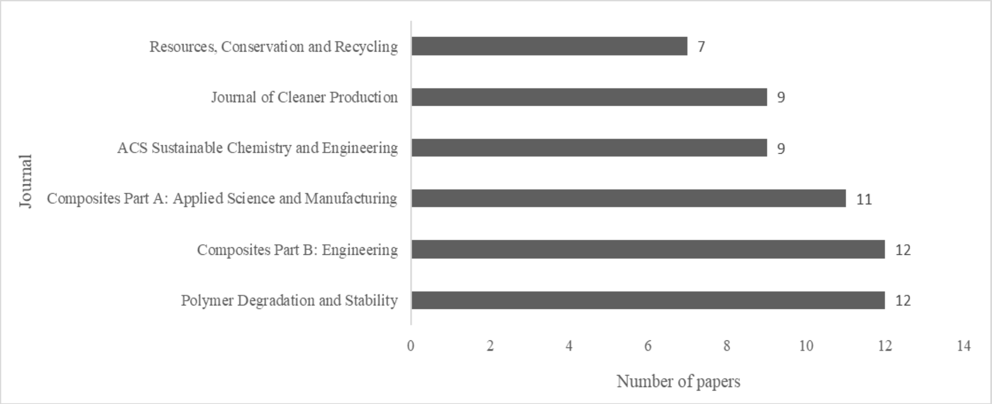

Figure 7: Top 6 Journals that publish on FRP recycling technologies

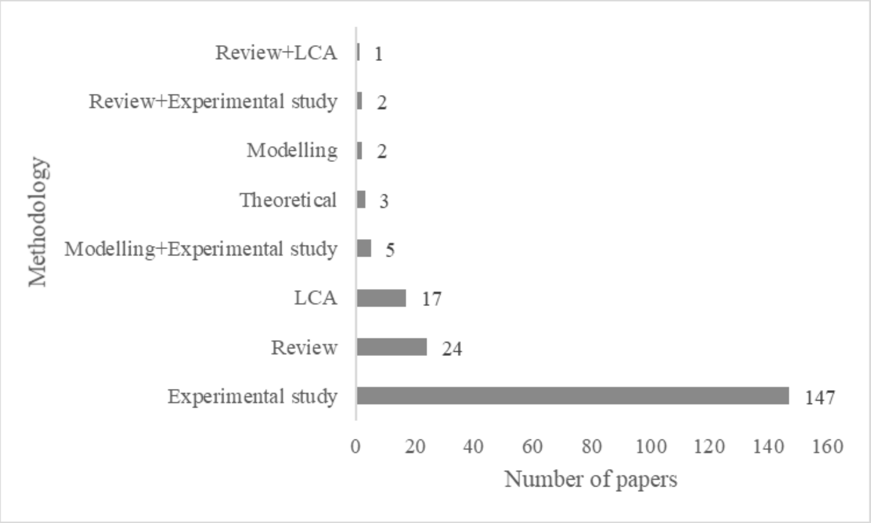

Figure 8: Type of research concerning FRP composite materials recycling technology

Experimental studies represent around 73% of the papers (147). Given the nature of the topic and the publications used as sources, this result is not surprising. In addition, 7 papers collaborate on an experimental investigation with another category, namely Modelling (5) and Review (2). The second type of article is a review article. These papers mostly review prior studies on FRP composite materials recycling technologies (Pimenta and Pinho, 2011), as well as provide comparisons between different recycling systems in select situations (Zhang et al., 2021). LCA is used in 17 studies that look at the environmental impact of various technologies (Vo Dong et al., 2018). All of the articles are about CF recovery, and the majority of them compare two or more recycling systems, or recycling technologies and/or landfilling and incineration. Only two articles use modelling, and there are only three works in the theoretical category. Cheng et al. (2019) used supercritical ethanol to simulate the degradation of a CFRP product under various conditions, while Hamel et al. (2020) developed a model to investigate the effect of reinforcement on the decomposition process of a CFRP with epoxy resin. Furthermore, Hagnell and Åkermo (2019) suggested a recycled material value model based on mechanical properties of recovered fibres, while Perry et al. (2014) claimed that designers and recyclers must share skills and knowledge to develop robust recycling processes. Overall, the fact that the majority of the studies are experimental studies appears to indicate that research in the examined area is still in its early stages (Colombo et al., 2021). Indeed, investigations primarily focus on analysing the mechanical properties of recoverable fibres using a unique recycling technology in order to determine their quality.

2.3.2.2 Science mapping

2.3.2.2.1 Keyword co-occurrence analysis

The VOSviewer output is a network with 19 nodes and 115 connections (Figure 9). Within the topic of FRP recycling technologies, this is a picture of the major of research terms and their co-occurrence correlations. It shows the most frequently used keywords and the strength of the links based on their co-occurrence. In general, the larger the node, the more frequently the considered keyword appears. As a result, the stronger the correlation between two specific keywords is, the stronger the link between the regarded objects is (van Eck and Waltman, 2010). Furthermore, the total link strength (TLS) measure represents the total strength of a node's links with other nodes (Hu et al., 2019; Ren et al., 2020). *Recycling* is the most prevalent keyword (133 occurrences), followed by *carbon fibre* and *carbon fibre-reinforced composites* (64 and 56 occurrences, respectively), *fibre-reinforced plastics* (47) and *chemical recycling* (47 occurrences) (35). These represent the current research hotspots. Since they were included in the query for detecting the papers to analyse, both *recycling* and *fibre-reinforced plastics* have significant positions. *Carbon fibre*, on the other hand, is a highly valuable raw resource in both economic and technical terms. As a result, it's unsurprising that research has centred on it rather than GF to build a system that enables for its successful recovery. Many *carbon fibre-reinforced composites* are nearing the end of their useful lives; therefore, it's critical to focus on recycling this type of material. *Chemical recycling* eventually arises, owing to the high quality of the fibres obtained by this recycling method. All of these keywords have the highest TLS values. This outcome indicates that these nodes are very important and central in the network. Thus, they are critical for developing research themes and bridging knowledge gaps.

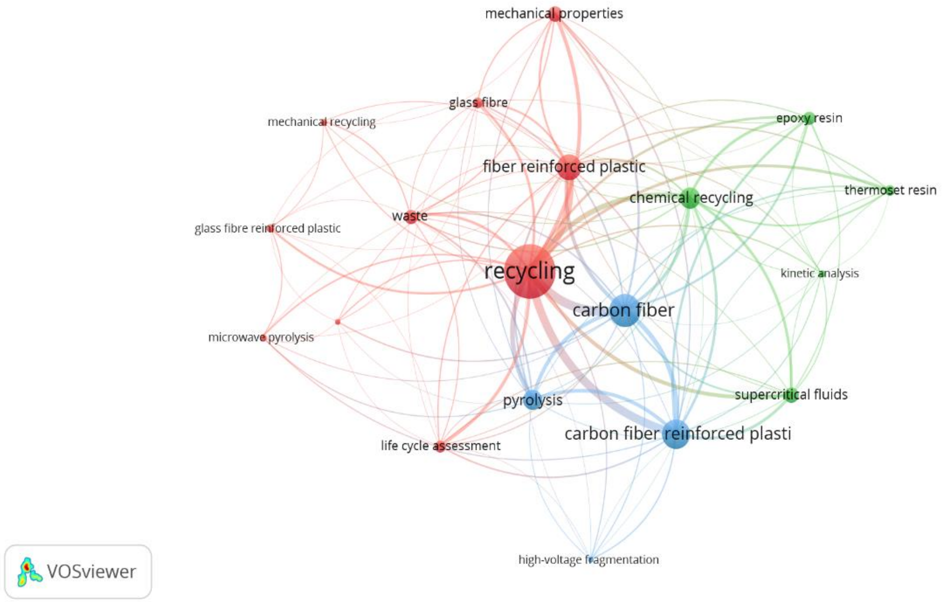

Figure 9: Authors' keywords network analysis

Cluster analysis

Three clusters were observed. Without a doubt, each cluster has some degree of relationship to a distinct recycling method. Cluster 1 encompasses 3 items, most of which are about mechanical recycling. The most notable exception is *microwave pyrolysis*, which is a thermal recycling process according to the literature. Since this keyword was linked to GFs and GFRPs multiple times (Åkesson et al., 2012; Kamimura et al., 2011), it is likely that it was included in this cluster. Five items belong to Cluster 2 and are heavily focused on chemical recycling. Within this group, supercritical fluids emerge as the most researched form of solvent for dissolving thermoset resins, particularly epoxy resins. Lastly, Cluster 3, which refers to thermal recycling, is the smallest, with 4 items. Indeed, such keywords shed light on the recycling of CFRPs to extract CF. This group includes the recycling technology known as *high-voltage fragmentation*, which can be classified as mechanical recycling, chemical recycling, or another recycling technology (Gopalraj and Kärki, 2020; Pakdel et al., 2020; Zhang et al., 2020). This outcome is probably a result of the fact that it has been used to recycle CFRPs in every study analysed. Each keyword's frequency and TLS are reported in Table 2.

Table 2: Information about the keywords for the three clusters

Cluster	Keyword	Occurrence	Total link strength
1	Recycling	133	293
	Fibre-reinforced plastic	47	111
	Mechanical properties	22	51
	Waste	20	47
	Life-cycle assessment	15	39
	Glass fibre	14	37
	Glass fibre-reinforced plastic	9	19
	Mechanical recycling	8	14
	Microwave pyrolysis	7	17
	Circular economy	6	16
2	Chemical recycling	35	101
	Supercritical fluids	22	62
	Epoxy resin	17	50
	Thermoset resin	14	36
	Kinetic analysis	7	22
3	Carbon fibre	64	164
	Carbon fibre-reinforced plastic	56	137
	Pyrolysis	29	75
	High-voltage fragmentation	6	17

The strongest relationships are found between *recycling* and *carbon fibre* (51), *recycling* and *carbon-fibre reinforced plastic* (46), *recycling* and *fibre-reinforced composites* (40) and *recycling* and *pyrolysis* (19). Moreover, following the removal of the keywords *recycling* and *fibre-reinforced plastic*, which would have distorted the analysis, Table 3 displays the links between the three groups. Looking at the values, some considerations can be drawn. The first point highlights the interconnectedness between these two macro-categories by showing how closely thermal and chemical recycling appear to be related to one another. This rise in research entropy (Miranda et al., 2019) witnesses the possibility that thermal and chemical recycling could soon merge into a single area of study. Additionally, both have ties to mechanical recycling. This finding demonstrates that mechanical recycling is a necessary prerequisite before thermal and chemical ones may be carried out, nowadays. However, thermal recycling has a stronger connection to mechanical recycling than chemical recycling. Indeed, thermal recycling, which includes pyrolysis that is used at an industrial level, is a more mature technology (Colombo et al., 2021). In such a context, a preliminary composite crushing step is probably needed.

Table 3: Keywords' co-occurrence between clusters

	Mechanical recycling	Chemical recycling	Thermal recycling
Mechanical recycling	64	24	57
Chemical recycling	24	86	76
Thermal recycling	57	76	112

In accordance with the method proposed by Bigliardi et al. (2021), Figure 10 shows an overview of the major study areas related to FRP recycling as well as a categorization of research topics based on the significance to academics. "Well-known" subjects include words like *recycling*, *fibre-reinforced plastic*, *carbon fibre*, *carbon fibre-reinforced plastic*, *chemical recycling* and *pyrolysis*, which are characterised by high incidence and

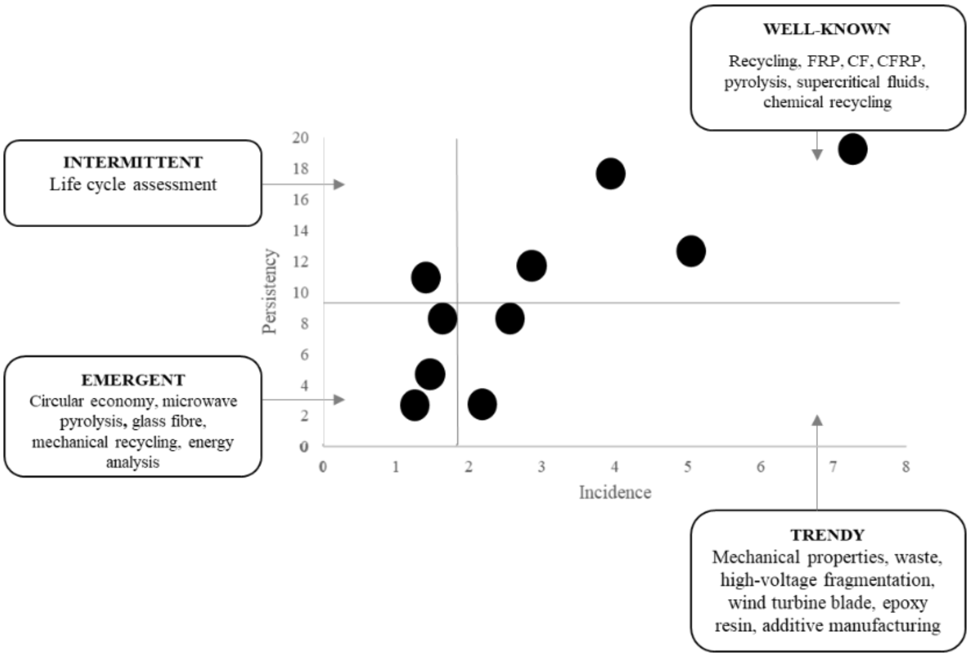

Figure 10: Authors’ keywords distribution considering Incidence and Persistency

high persistence. Due to the fact that they represent incredibly wide and comprehensive notions, these keywords are the most frequently used by academics. Even if the term *pyrolysis* refers to a particular technology, it can be included in this group because it is used commercially (Pakdel et al., 2020; Pimenta and Pinho, 2011). Instead, "emerging" topics are those that have the potential to become trendy or rapidly disappear. It encompasses terms such as *microwave pyrolysis*, *energy analysis* and *circular economy*, which are intriguing subjects because they have just recently, within the past ten years, been employed. Therefore, in the upcoming years, researchers should concentrate on these issues. Finally, this category includes mechanical recycling. Currently, it is nearly always mentioned in the context of the development of other technologies (e.g. LCA of mechanical recycling as a comparison target). This fact also demonstrates that, barring the development of disruptive technologies, mechanical recycling research has reached its technological limit. As a result, it may be referred to as a phantom topic. *High-voltage fragmentation* is a keyword that falls under the "trendy" category, namely among subjects that academics working in this area of study seem to think have a lot of potential. In fact, it permits the recovery of clean and long fibres from CFRPs (Oshima et al., 2020) without having a significant negative influence on the environment. Within this group, there are also other particular terms like *additive manufacturing* and *wind turbine blade* that now have few citations and, consequently, a limited number of occurrences. Lastly, “intermittent” topics are researched in a discontinuous way. Life-cycle assessment is the sole keyword included in this group. Since 2013, several viewpoints have been taken into account when researching this topic, however the majority of studies belong to the year 2021 (e.g. Zubail et al., 2021). For instance, Nunes et al. (2018) conducted an LCA analysis to determine the effects of steam thermolysis recycling of CFRP waste on the environment. Different recycling scenarios, such as landfilling and incineration with pyrolysis, solvolysis and mechanical recycling, were instead analysed by other scholars, who either evaluated the scenarios separately or in combination (Fanran Meng et al., 2018; Pakdel et al., 2020). This outcome can be explained by the fact that a technology needs to be properly established prior to an environmental analysis. Furthermore, as was already noted, sustainability is a subject that is quite susceptible to external pressure. Therefore, significant expansion in this area is foreseen in the upcoming years since environmental impact analysis is a component of the strategies of the world's economies.

The usage patterns of terms throughout time are not taken into consideration by a keyword co-occurrence map, which is a static picture (Si et al., 2019). By examining papers from the years 2007 to 2021, the researcher was able to bridge this gap. The decision to focus on these years was made since, starting in 2007, there has been a noticeable increase in the quantity of publications. Papers were divided into two time periods, specifically 2007-2015 (38 papers) and 2016-2021 (131 papers). Indeed, in 2016 there was a further significant rise in the number of publications published. The authors were able to look at the most recent study trends by setting these parameters. The keyword co-occurrence bibliometric maps for the time periods 2007-2015 and 2016-2020, respectively, are shown in Figure 11 and Figure 12. To establish the best balance between the readability and completeness of the map, the minimum number of occurrences of a specific keyword was set to two for the first period and six for the second. In the first instance, 17 out of 26 of the keywords met the requirement, while in the second, 17 out of 39 did. The TLS and keyword occurrences for the two time periods under investigation are shown in Table 4, with new terms that emerged in the second period highlighted in italics.

All in all, interest in sustainability-related themes has grown in recent years. Indeed, *Circular economy* can only be seen in the second period. This outcome is consistent with the fact that the Ellen MacArthur Foundation first used this expression in 2013 (Ellen MacArthur Foundation, 2013). It also emphasizes the significance of a thorough examination of circular economy practices within this research topic. Studies should actually concentrate on minimizing waste and manufacturing leftovers, as well as reuse, waste reduction and recycling as a practice. *High-voltage fragmentation*, the other new word that appeared in the second period, alluded to the growing significance of environmental issues. Furthermore, since the minimum number of occurrences was set to six, it is important to note that the keyword *microwave pyrolysis* is no longer present in the second period under investigation. Such a finding demonstrates, in light of the preceding analysis, that this issue cannot be regarded as phantom but rather as a niche topic with some room for expansion as it exhibits the occurrence of three (not far from that of the previous period).

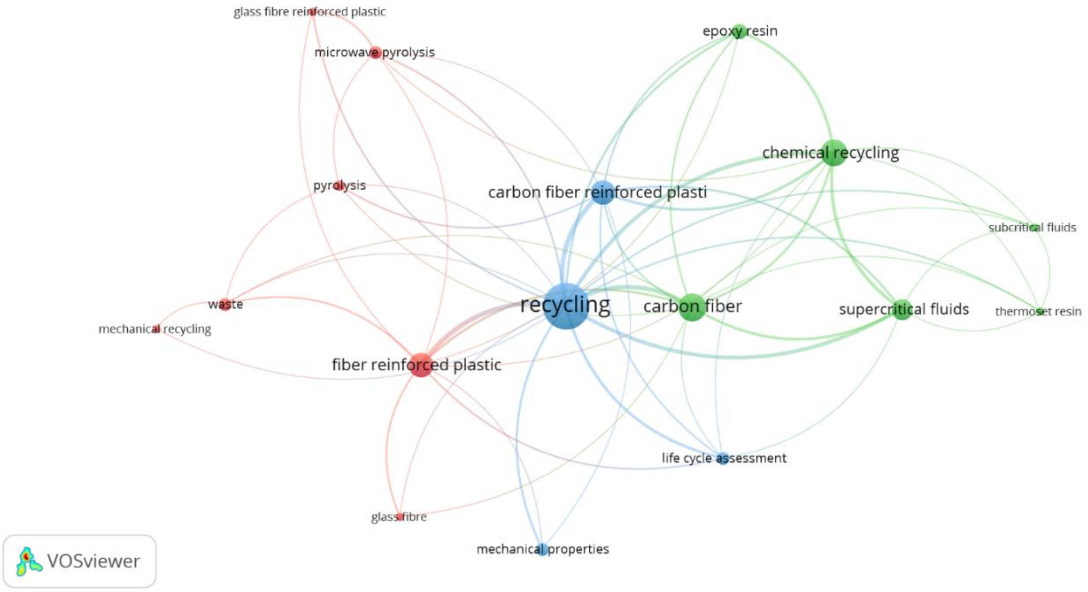

Figure 11: Author's keyword map based on publishing year (time period: 2016-2021)

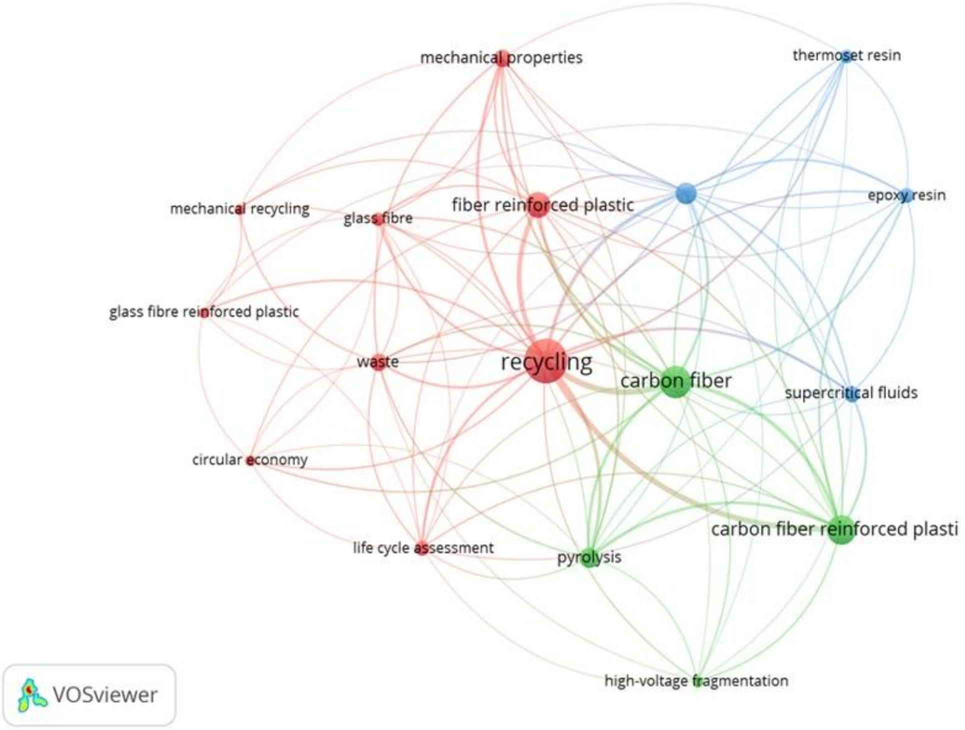

Figure 12: Author's keywords map based on publishing year (time period: 2007-2015)

Table 4: Main keywords with occurrences and total link strength

First period: 2007–2015				Second period: 2016–2021			
#	Item	Occurrence	TLS	#	Item	Occurrence	TLS
1	Recycling	25	61	1	Recycling	99	229
2	Fibre-reinforced plastic	16	40	2	Carbon fibre	52	141
3	Carbon fibre	12	31	3	Carbon fibre-reinforced plastic	45	109
4	Chemical recycling	11	29	4	Fibre-reinforced plastic	45	112
5	Carbon fibre-reinforced plastic	10	26	5	Chemical recycling	23	77
6	Supercritical fluids	8	24	6	Pyrolysis	22	60
7	Epoxy resin	5	13	7	Mechanical properties	17	45
8	Life-cycle assessment	4	12	8	Waste	15	40
9	Microwave pyrolysis	4	8	9	Supercritical fluids	14	39
10	Pyrolysis	3	6	10	Epoxy resin	12	38
11	Subcritical fluids	2	7	11	Life-cycle assessment	11	28
12	Thermoset resins	2	6	12	Thermoset resins	10	28
13	Waste	4	6	13	Glass fibre	9	28
14	Glass fibre-reinforced plastic	2	5	14	*High-voltage fragmentation*	6	17
15	Mechanical properties	4	6	15	*Circular economy*	6	15
16	Glass fibre	2	4	16	Glass fibre-reinforced plastic	6	12
17	Mechanical recycling	2	2	17	Mechanical recycling	6	12

2.3.2.2.2 Document co-citation analysis

The network of document co-citations is shown in Figure 13. The fact that there were only four publications published before 2007 emphasizes how new the subject at hand is. Table 5 provides details on the studies that have had the most influence in the subject of FRP recycling technology. Overall, there are not many co-citations of documents (89 citations for the top-ranked paper and 25 citations for the last ranked). This result may be probably traced back to the relatively tiny size of the document database under consideration. Moreover, the analysis made it possible to identify two separate clusters. Ten documents from Cluster 1 are concerned with conventional recycling techniques. Instead, Cluster 2 consists of seven articles that are directly related to supercritical fluids and chemical recycling. The first group's papers were all published before 2016, which is when the second growing wave of publications (2016-2020) began, with the exception of

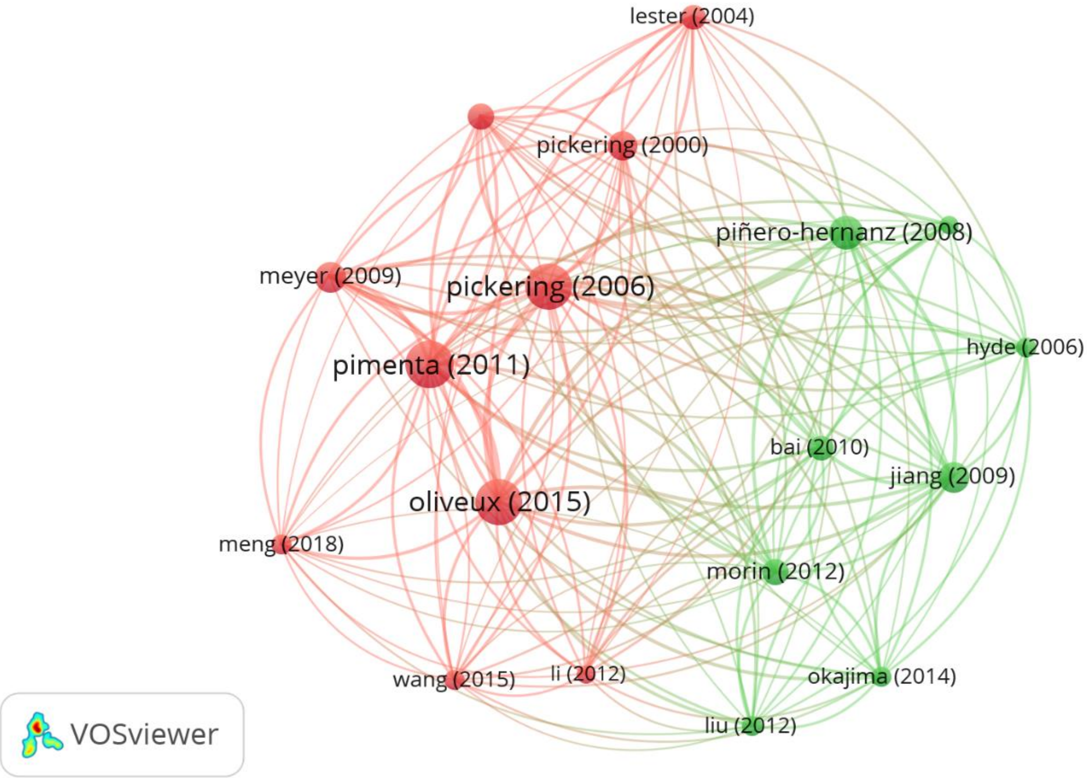

Figure 13: Co-citation map of cited references

one. The top three documents in terms of citation and TLS rank among them (Oliveux et al., 2015; Pickering, 2006; Pimenta and Pinho, 2011). In particular, they are review papers with various objectives. They undoubtedly had, and continue to have, a significant impact on the field owing to their comprehensiveness. Pickering (2006) divided thermosetting composite material recycling technologies into mechanical, thermal and chemical macro-categories. Instead, Pimenta and Pinho (2011) attempted to summarize the most recent advancements in CF recovery and remanufacturing technologies as well as potential uses for final goods composed of recovered materials. Finally, Oliveux et al. (2015) employed LCA to focus their attention on both the technical and economic and environmental aspects of recycling technologies for composite materials reinforced with both CFs and GFs.

Table 5: Information about the papers with the highest impact in the FRP recycling technologies field

Cluster	Paper	Title	Citation	TLS
1	Pimenta and Pinho (2011)	Recycling carbon fibre reinforced polymers for structural applications: Technology review and market outlook	89	451
1	Pickering (2006)	Recycling technologies for thermoset composite materials -current status	86	463
1	Oliveux et al. (2015)	Current status of recycling of fibre reinforced polymers: Review of technologies, reuse and resulting properties	85	478
1	Meyer et al. (2009)	CFRP-recycling following a pyrolysis route: Process optimisation and potentials	49	294
1	Pickering et al. (2000)	A fluidised bed process for the recovery of glass fibres from scrap thermoset composites	45	266
1	Palmer et al. (2009)	Successful closed-loop recycling of thermoset composites	40	266
1	Lester et al. (2004)	Microwave heating as a means for carbon fibre recovery from polymer composites: A technical feasibility study	36	223
1	Wang et al. (2015)	Chemical recycling of carbon fiber reinforced epoxy resin composites via selective cleavage of carbon-nitrogen bond	29	180
1	Meng et al. (2018)	Comparing life cycle energy and global warming potential of carbon fiber composite recycling technologies and waste management options	27	170
1	Li et al. (2012)	A promising strategy for chemical recycling of carbon fiber/thermoset composites: Self-accelerating decomposition in a mild oxidative system	25	194
2	Piñero-Hernanz et al. (2008)	Chemical recycling of carbon fibre reinforced composites in nearcritical and supercritical water	58	378
2	Jiang et al. (2009)	Characterisation of carbon fibres recycled from carbon fibre/epoxy resin composites using supercritical n-propanol	49	339
2	Morin et al. (2012)	Near- and supercritical solvolysis of carbon fibre reinforced polymers (CFRPs) for recycling carbon fibers as a valuable resource: State of the art	40	271
2	Bai et al. (2010)	Chemical recycling of carbon fibres reinforced epoxy resin composites in oxygen in supercritical water	37	281
2	Okajima et al. (2014)	Chemical recycling of carbon fiber reinforced plastic using supercritical methanol	29	226
2	Liu et al. (2012)	Chemical recycling of carbon fibre reinforced epoxy resin composites in subcritical water: Synergistic effect of phenol and KOH on the decomposition efficiency	27	206
2	Liu et al. (2004)	Recycling of carbon/epoxy composites	26	175

This new and unique viewpoint emphasizes the growing significance of the sustainability as a whole. Similarly, Fanran Meng et al. (2018) assessed the environmental impact of CFRP recycling technologies by comparing them simultaneously in a more recent study. In detail, they looked at landfill, incineration, mechanical recycling, fluidized bed, pyrolysis and chemical recycling in the UK context. Overall findings indicate that when compared to landfill and incineration, recycling scenarios are frequently the most environmentally favourable choices. The remainder of the documents all discuss a particular recycling technology, giving details on both its technical application and its experimental outcomes. Specifically, the oldest articles focus on the analysis of mechanical and thermal recycling technologies (Lester et al., 2004). The most recent ones, on the other hand, concentrate on the analysis of various chemical recycling technologies. For instance, Li et al. (2012) studied an effective hydrogen peroxide-based method for recycling CFRPs made of epoxy resins (recognised as a good oxidant due to its environmentally friendly nature). Instead, Wang et al. (2015) used acetic acid to their advantage, which by swelling the dense epoxy resin structures enables recovery of both oligomers and CFs.

Regarding Cluster 2, all publications with the exception of one were published during the initial period under investigation. In addition, all of them are experimental studies, with the exception of Morin et al. (2012), who summarize recycling technologies focused on solvolysis by near- and supercritical fluids. This finding demonstrates that basic research on chemical recycling has been conducted from the beginning of the study period. To address the sustainability issue, research has recently moved its attention to greener solvents. Overall, it is a broad macro-category that encompasses a number of technologies distinguished by various characteristics that require thorough research. Over the past few years, scientific interest has primarily shifted to the study of solvolysis via supercritical fluids. Indeed, they are good solvents, suitable for the advancement of environmentally friendly recycling technologies. Water in particular, which is “cheap, recyclable, non-toxic and reasonably easy to manage” (Liu et al., 2012) fits this description. In accordance with this pattern, the paper of Piñero-Hernanz et al. (2008) has received the most citations and has the highest TLS value within this group. This article has a considerable influence on the field; indeed, it is the fourth most commonly co-cited document in the whole database. The authors of this study demonstrated that CFs recycled without the use of an alkali catalyst have good mechanical properties by investigating the solvolysis of the epoxy resin from CFRPs in supercritical water conditions. The recycling of CFs from CFRPs by using supercritical water with an excess of oxygen was also suggested by Bai et al. (2010). Recycled CFs exhibited excellent surface and mechanical properties without overoxidation. Instead, phenol and potassium hydroxide were combined in subcritical water by Liu et al. (2012). As a result, the CFs produced had a higher tensile strength than the reaction system using only potassium hydroxide. Other solvent types have also been examined. Jiang et al. (2009), for example, looked into the possibility of recycling CFRPs using supercritical n-propanol. This fluid was chosen because it can be processed using more friendly processing conditions than supercritical water. Despite maintaining their mechanical properties, the new epoxy resin significantly decreased the interfacial bonding strength. Besides, Okajima et al. (2017) investigated supercritical methanol. Finally, nitric acid solutions were studied a while back. By using a solvolysis at a low temperature, Liu et al. (2004) discovered regenerated CFs with suitable mechanical characteristics. Despite this favourable conclusion, this type of solvent has been gradually phased out due to its aggressiveness and potential environmental hazards.

2.3.3 Future research avenues

The results of the bibliometric analysis were further examined in order to draw some conclusions and identify possible research gaps that could be turned into potential directions for future studies.

First, there has been an increase in the research into recycling technologies for FRP composite materials over time. Indeed, after ten years marked by low publishing productivity, interest in the subject has significantly increased since 2012, particularly in the previous five years. Therefore, it is conceivable to assume that the number of articles published in the near future will grow gradually and continuously, signalling an increase in research effort. The pressure from regulation and the growing market knowledge of the sustainability issue might be attributed to this tendency (Dyllick and Muff, 2016). Second, the most popular methodology for research is the experimental study, highlighting the fact that the field of study is still developing (Colombo et al., 2021). In the case of chemical and thermal recycling, this result is especially accurate. Particularly, works on chemical recycling receive the greatest citations and constitute a cluster on their own.

Such papers were composed throughout the first time period taken into account for the temporal analysis of the keywords, demonstrating the persistently high level of interest in this subject. This does not mean that experimental research on chemical recycling is finished, though. Indeed, due to the growing emphasis on sustainability and upcoming legislation, researchers are currently concentrating on the investigation of solvents with a lower environmental impact (e.g. Lee et al., 2020).

By means of the keyword analysis, a gap was discovered regarding the lack of focus on the usage of recycled fibres in end applications. Indeed, there are no keywords associated with that issue in Figure 9. Therefore, one direction for future study is to assess the feasibility of manufacturing methods for FRP composite materials adopting recycled fibres for certain final applications, in addition to recycling technologies. In fact, that feature should be considered when developing novel recycling technologies or improving ones that already exist. Similarly, the "market characteristics" of FRP composite materials, such as the availability of manufacturing scraps or end-of-life waste, or a channel for the purchase and sale of products made of recycled fibres, as well as the supply chain for material collection for recycling, may also be the subject of further research.

Concerning chemical recycling, it is reasonable to argue that, despite its beneficial effects on the quality of recovered fibres, solvolysis by strong acids is no longer feasible due to environmental restrictions. To get around this restriction, academics have been concentrating on researching new solvents and/or technologies in recent years. Despite requiring more energy consumption, using supercritical water alone or in combination with green solvents is still a good solution, according to the study of the pool of papers. However, it is only utilized in laboratories to date (Gopalraj and Kärki, 2020). Additionally, electrochemical recycling was identified as an alternative technique. This sounds like a good choice because it is efficient, affordable and simple to use. Accordingly, research should continue to concentrate on supercritical fluids in order to industrialise the technologies that employ them and focus on environmentally friendly technologies. The study of pyrolysis has grown throughout time. This resulted in the industrialisation of the process, which enabled its commercial viability. However, putting it into practice takes a lot of energy. Microwave pyrolysis has been developed as a solution to this problem. With such technology, the process time is reduced in addition to the need for lower temperatures. However, the analysis indicates that it is a niche subject. An increase in the study effort for this technology is expected, given the growing relevance of the sustainability issue. Instead, since mechanical recycling severely decreases the mechanical properties of the fibres, it garnered much less attention than other recycling technologies, and academics' interest in it has waned over time. Nowadays, it is mostly used to reduce the size of FRP composite materials waste as a pre-recycling procedure for more sophisticated recycling technologies. High-voltage fragmentation seems to be a practical and sustainable method today for reducing waste and recovering long, spotless fibres. Given these benefits, this solution unquestionably deserved additional research.

Overall, the study areas identified as "emerging" and "trendy" through the keyword analysis, such as the circular economy, microwave pyrolysis, high-voltage fragmentation and additive manufacturing, should receive special attention due to their current significance. Specifically, to reuse recovered fibres from CFRP waste for new structural applications, additive manufacturing may present a possible strategy (Huang et al., 2020; Liu et al., 2021), strengthening the connection between the circular economy and recycling technologies (Clemon and Zohdi, 2018).

Last but not least, highlighting the significance of LCA studies is also helpful. They do, in fact, make it possible to compare various recycling choices with one another or with landfilling and incineration, revealing information on the environmental sustainability of each recycling technology. Meanwhile, two crucial issues have not yet been comprehensively addressed in the literature. The first relates to economic analysis, which has only rarely been combined with LCA in research. The economic evaluation should be further looked at as it is essential to comprehend the economic feasibility of FRP composite recycling technologies in order to advance them to the industrial level (Vo Dong et al., 2018). The social sphere is the subject of the second aspect. Indeed, Figure 9 does not show any keywords related to social behaviour or acceptability. This outcome implies that this component is still insufficiently developed to manifest. However, it is crucial to define and describe the effects of new or established recycling technology on society and human health (Khalil, 2018). Considering the rising interest in both the sustainability issue and the CE principles, both of these issues merit further investigations.

2.3.4 Limitations and research improvements

The bibliometric analysis performed has some limitations. First, the bias of the researchers inevitably has an impact on the systematization of the data. This problem was lessened by explaining the research procedure as transparently as possible. Additionally, it should be mentioned that the content was only gathered using the Scopus database. It could be beneficial to conduct research using databases other than this one, such as Web of Science or Google Scholar, to confirm the results of this analysis. Furthermore, the analysis has only considered scientific publications. Grey literature inclusion might lead to significant contributions. These limitations obviously give a chance that could be exploited in upcoming works.

2.4 Patent analysis

2.4.1 Methodology

A patent technology roadmap was used to enhance knowledge about prior, current and future use of the main FRP composite materials recycling technologies and, accordingly, shed light on the actual technology maturity level. To this end, three different types of analysis were carried out. In detail, a time series analysis was performed to emphasize the maturity level of existing mechanical, thermal and chemical recycling methods, predict their expected remaining life and evaluate their potential of growth. Through a patent trend analysis was possible to identify the main industries affected in the topic at hand and to investigate how their interest would change over time. Lastly, citation analysis was used to understand whether there were any links or interdependencies between existing recycling methods. The following is a full discussion of each approach.

2.4.1.1 Time series analysis

The growth/decline rate of a technology over time was examined using time series analysis (Widodo et al., 2011). In detail, a Gompertz growth curve model was used, as proposed by Yoon et al. (2014). The total number of patents filed between 1991 and 2018 served as the input data. This was achieved since an S-shaped curve may be used to describe the life cycle of a technology, whose development tendency can be approximated by the number of patents gathered over time (Cioffi, 2005). The equation that defines the Gompertz model is as follows:

$$Y_t = Le^{-ae^{-bt}} \quad (1)$$

where L is the asymptotic maximum value (i.e. saturation level) attained by Y_t, the model’s dependent variable (i.e. the cumulative number of patents belonging to a single macro-category at time t) and a and b are the curve's location and shape, respectively.

A least square estimation method was adopted to determine the values of the three parameters (Yoon et al., 2014). Furthermore, the online Loglet Lab program[1] was used to execute computations and, as a result, fit the obtained data. The software allows the researcher to verify the statistical significance of the data and automatically construct an S-shaped curve by processing the curve fitting. Moreover, it enables the researcher to determine the stage at which each technology has reached in its life cycle, which, according to Haupt et al. (2007) and Mann (2002), can take one of four forms: birth, growth, maturity, or saturation. Each configuration is distinguished by different shares of upper limits (Ernst, 1997), as reported in Table 6.

Table 6: Technology life cycle stages by upper limit share (authors' elaboration of Ranaei et al. (2014))

Share of Upper Limit	Stage of Technology Life Cycle
$Yt/L \leq 10\%$	Birth
$10\% < Yt/L \leq 50\%$	Growth
$50\% < Yt/L < 90\%$	Maturity
$Yt/L \geq 90\%$	Saturation

2.4.1.2 Patent trend analysis

To achieve the study's second goal, a patent trend analysis was performed. First, the assignees, or patent owners, were classified into two categories: public and private sector organizations and individuals. The first category was further separated into two subgroups: research centres and universities in the first, and private companies in the second. Then, using the Bureau van Dijk's Orbis[2] database, private organizations were classified using their NACE code (Revision 2) as a guide. Second, each assignee was given a score ranging from 0 to 1 based on their percentage ownership of the patent in question: for example, if there was just one owner, the value allocated was 1, but if there

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[1] https://logletlab.com/

[2] https://orbis.bvdinfo.com/

were two owners, it was 0.5. This assumption was made to avoid overestimating the ownership of patents. Third, the interest of each sector (i.e. NACE codes, Research and Individuals) was determined by adding up the estimated scores. Lastly, a year-by-year stratification was carried out to investigate the potential change in interest over time.

2.4.1.3 Citation analysis

The backward citations in the cited patents were used to build the citation analysis. As backward citations provide information regarding an invention's technological antecedents (Jaffe and De Rassenfosse, 2019), they were considered particularly ideal for attaining the study's third goal. Each patent in our database (i.e. citing patents) had its backward citations (i.e. cited patents) classified according to whether it belonged to one of three main recycling macro-categories: mechanical, thermal, or chemical. The citations that did not fit into any of the above categories were assigned to a fourth macro-category, called 'others'. As a result, the cited patents’ publication numbers were compared to the publication numbers of the patents in the whole dataset. An interdependence index was generated by comparing these groups to the overall number of backward citations in the respective macro-category. The degree of citation inside and between each macro-category is measured by this index.

2.4.1.4 Procedure of the research

Overall, the workflow of the study involves five stages: database design, search strategy definition, patent pool retrieval, patent analysis and results discussion. The search database was defined in the first stage. Data were collected utilizing Questel Orbit Company's Intellectual Property Portal[3], which is a platform that provides data mining tools to examine individual databases with unique attributes, such as legal status. Orbit is also built on patent families, which are collections of granted patents and patent applications that protect the same idea (*OECD Pat. Stat. Man.*, 2009). This prevents duplication of patent document analysis, which is common when corporations patent the same technology in numerous countries (Karvonen et al., 2016). The second stage entailed determining the search technique. To circumvent the constraints of both the keywords method and the classification code strategy, a mixed strategy was used. First

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[3] https://orbit.com/,

and foremost, a keyword search was conducted. Keywords relevant to the FRP composite material recycling methods domain were inputted in the English language, including all potential variants, then searched in the patent record's title, abstract and claims fields (Table 7). At the same time, an attempt was made to locate the appropriate International Patent Classification (IPC) codes. To do so, a categorization code tree was combed through, followed by a reverse engineering examination to find the IPC codes that were essential to the investigation.

Table 7: Keywords for constructing the overall patent pool

Search keywords	
Overall patent pool	Fibre reinforced plastic, Fibre reinforced polymer, GFRP, CFRP, Recycling, Recovering, Reclaiming

The results of the two assessments were merged, and a committee of specialists in the subject was brought in to refine the sample of patents that had been chosen. Despite the fact that it was time demanding, this exercise enabled the researcher to identify the words that were causing noise in the sample. Furthermore, following the manual screening, an algorithm based on keyword features, proximity in the document, and recurrence was used to reduce the remaining noise. Afterwards, using defined keywords (Table 8), this sample was sorted into three recycling macro-categories (mechanical, thermal and chemical).

Table 8: Keywords for constructing the patent pool of each recycling technology

Search keywords	
Mechanical recycling	Mechanical, Pulverizing, Powdering, Grinding, Crushing, Shredding
Thermal recycling	Thermal, Thermic, Pyrolysis, Fluidized bed
Chemical recycling	Chemical, Solvolysis, Hydrolysis, Glycolysis, Acid, Solvent

There were 205 patent families for mechanical recycling methods, 389 for thermal recycling processes and 494 for chemical recycling procedures among the three samples. The patent analysis was carried out in the fourth stage using a time series, patent trend and citation study. Lastly, a thorough discussion of the obtained results was carried out in the fifth stage.

All patents considered in this investigation had a priority date, which is the year in which the patent was first filed in a legal office, between 1991 and 2020. This decision was made since, prior to 1991, patent activity on this area was negligible when compared to the period of time studied. Due to the 18-month delay between filing and publication, the authors are also aware that the two-year period 2019-2020 is not indicative of the patterns at hand. Finally, the patents were gathered in December 2020.

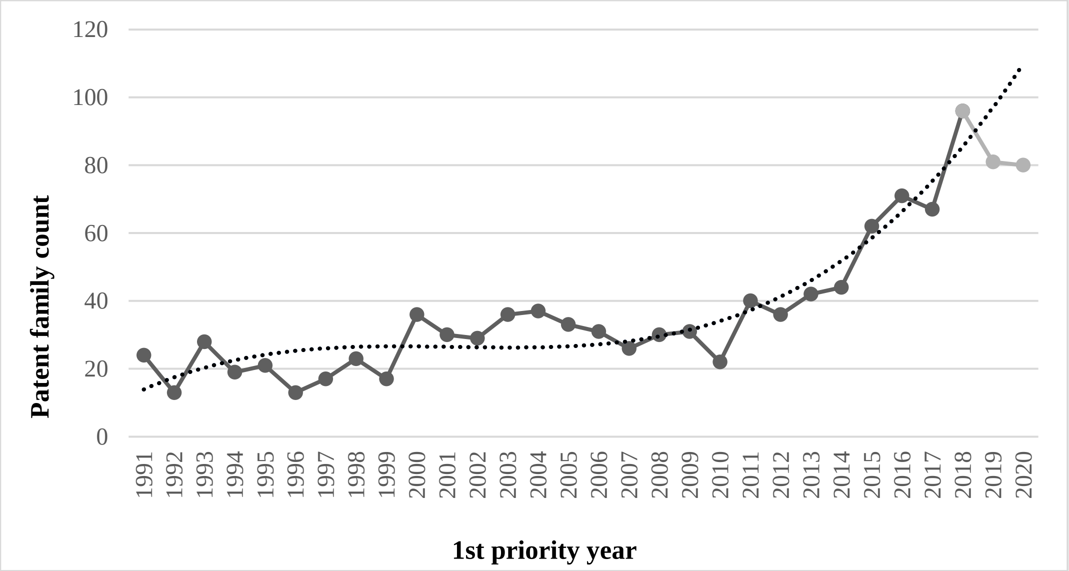


Figure 14: Historical trend of filed patents by first priority year (black line represents the trend of patents filed between 1991-2018, grey line represents the trend of patents filed during the non-representative two-year period 2019–2020, and dotted line represents the overall trendline)

2.4.2 Outcomes and related discussion

2.4.2.1 Time series analysis

As shown in Figure 14, patenting activity on FRP composite materials has been steadily increasing over the last 30 years, with the highest number of issued patents (82) in 2018. This trend reflects the growing interest in research in the development of innovative technologies for the recycling of FRP composite materials. Nonetheless, since the data is unreliable, the decline in patenting activity that occurred during the two-year period 2019–2020 cannot be considered vital for the study. Indeed, many patents filed during this period of time may not have yet been published, given the average time between filing and publication was 18 months. By analysing the historical trend of relevant patenting activity on a global scale, the significant interest of research in developing novel and effective recycling procedures for FRP composite materials can also be deduced (Figure 15).

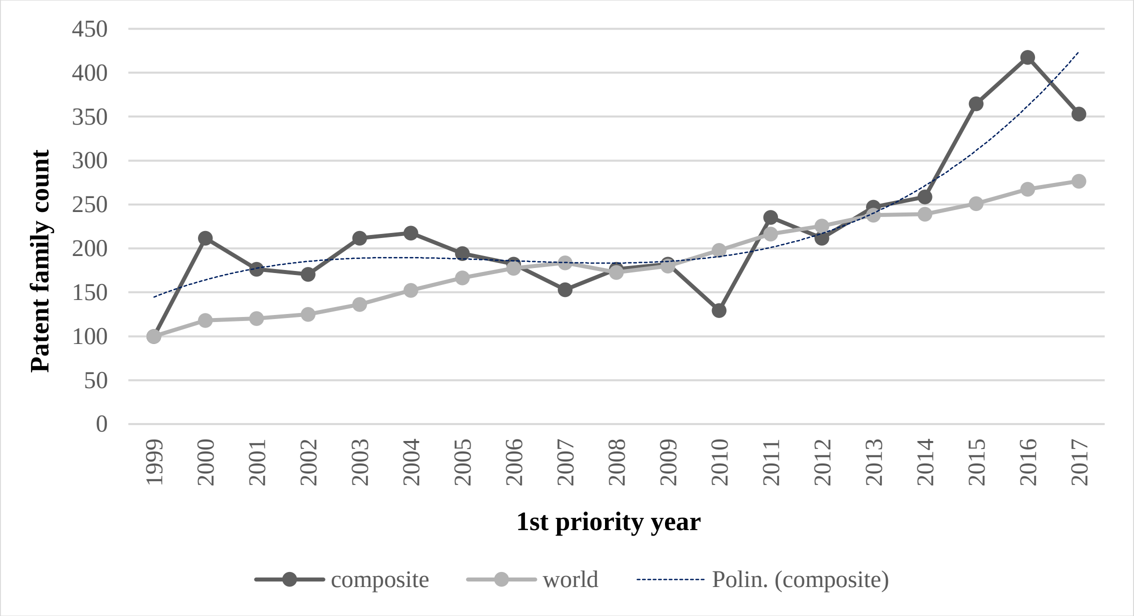


Figure 15: Benchmark between composite recycling technologies patent activity and worldwide patent activity

Furthermore, such a comparison reveals that patenting activity on recycling systems for FRP composite materials is increasing at a slightly higher rate than global patenting activity. The underlying motivation can be complex, and can be influenced by both internal and external causes. Indeed, environmental sustainability has been increasingly important in policy (Kumar et al., 2019). Additionally, society as a whole is becoming increasingly environmentally conscious (Brochado et al., 2017). Customers, whether B2B or B2C, are urged to acquire or have products that adhere to the principles of sustainability and circular economy for a variety of reasons, including market competitiveness and social standing (Ali et al., 2019; Miranda et al., 2019). Internal causes contribute to this rapid expansion, most notably the necessity for businesses to discover technology solutions that enhance production efficiency and, as a result, cut prices. Recycling, on the other hand, can help businesses become less vulnerable to market fluctuations. For instance, the ability to recycle larger amounts of CFs would allow companies to be less reliant on the price of virgin raw materials, whose value is heavily associated with the price of oil, which is volatile by nature (Ellringmann et al., 2016).

The FRP recycling technologies market is still in its early stages, as shown by the Gompertz growth curve in Figure 16 (technology maturity level (TML) = 36.57%).

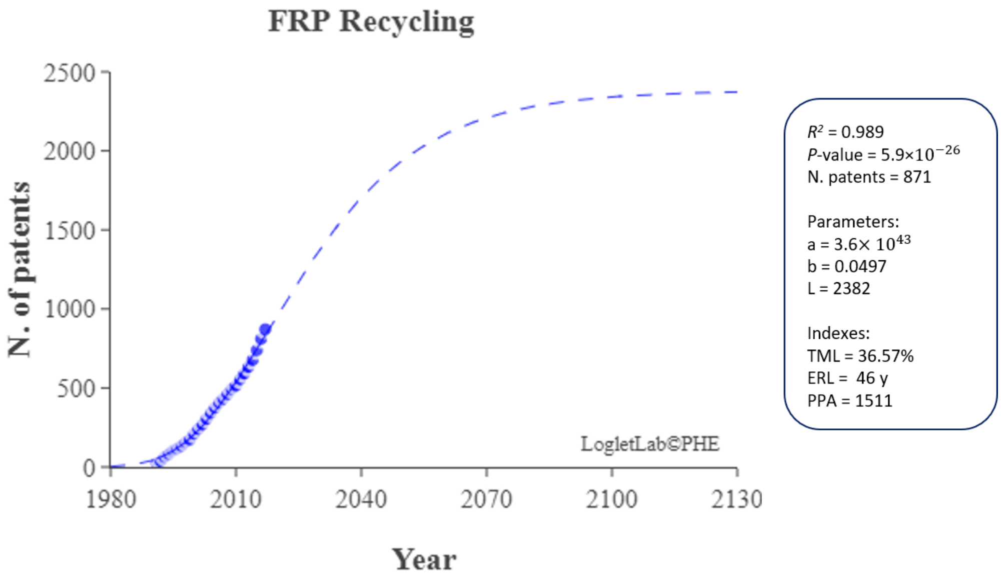


Figure 16: Gompertz growth curve and its parameters for FRP recycling

Nonetheless, considering the expected remaining life ((ERL) = 46 years), the number of patents is likely to continue on this upward trajectory for at least the next 10 years. Finally, the number of possible patents (PPA = 1,511) suggests that future patenting activity in this market will be higher than in the past, implying that academic and industrial researchers will continue to focus on the topic.

Figure 17 displays the evolution of patents filed on mechanical, thermal and chemical recycling technologies between 1991 and 2020. Since 2010, patenting activity has been steadily increasing in all three recycling macro-categories, according to the statistics. Instead, the Gompertz growth curve (Figure 18) shows that mechanical recycling technologies have reached maturity (TML = 61.51%), whereas thermal (TML = 36.54%) and chemical recycling technologies are still in the early stages of development. Chemical recycling technologies, in particular, have the lowest maturity level (TML = 30.96%). Furthermore, the findings suggest that chemical (ERL = 48 years) and thermal (ERL = 51 years) recycling technologies have an estimated remaining life that is roughly twice that of mechanical recycling (ERL = 24 years). In conclusion, the data analysis shows that when examined from the standpoint of the three recycling macro-categories, the progressive growth in the number of patents takes on diverse forms, indicating the existence of technologies that have matured faster than others. This behaviour could be attributable to each technology's complexity and history. Mechanical recycling of CFs, for instance, has risen more quickly than thermal and chemical recycling, as it is easier and is based on existing knowledge and technologies from other industries. Furthermore, owing to its advanced maturity, it is on the verge of reaching the technological upper limit of the technological transformation process, which is defined as the collection of technological actions required to recover the fibres. In the case of the technical transformation system, which is described as a set of processes, tools and equipment ideal for an effective, efficient and reliable recycling process, there is still plenty of potential for development. Thermal and chemical recycling, on the other hand, are still defined by prospects for growth and improvement from a technological point of view, as they are in the growth phase of their life cycle.

2.4.2.2 Patent trend analysis

Table 9 lists the most active players in the field, organized by recycling macro-category.

Table 9: Patent holdings by recycling macro-category and assignee category

Actors	Mechanical Recycling	Thermal Recycling	Chemical Recycling
Research	8.4%	15.0%	26.7%
Individuals	10.2%	9.8%	6.6%
Manufacture of chemicals and chemical products	14.8%	13.8%	22.5%
Manufacture of rubber and plastic products	6.4%	6.9%	4.0%
Manufacture of electrical equipment	3.8%	7.1%	13.0%
Manufacture of motor vehicles, trailers and semi-trailers	10.1%	7.1%	3.1%
Construction of buildings	6.6%	2.7%	-
Other	39.7%	37.6%	24.1%

First, the data shows that for each recycling macro-category, the top categories of assignees affected by the subject at hand are the same, indicating a thorough concentration of interest in the topic. Second, it's worth noting that 'Research' is the most significant domain for both chemical and thermal recycling, with 26.7% and 15.0%, respectively, whereas it is only fourth with 8.4% for mechanical recycling. These results demonstrate the technologies’ maturity level discussed in Section 2.4.2.1. Indeed, the development of chemical and thermal recycling technologies is still in its early stages, as evidenced by the large number of patents developed at the research level.

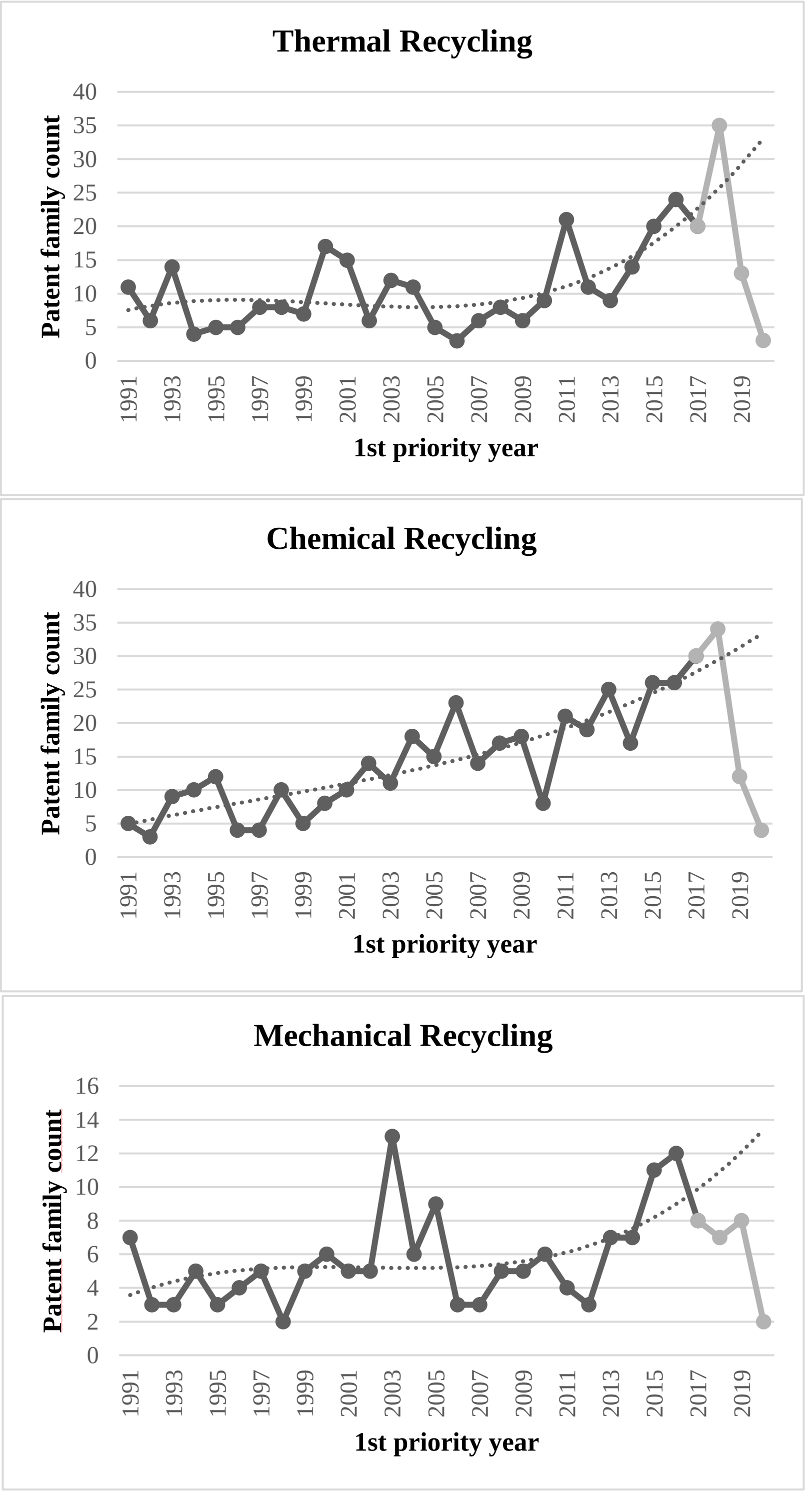
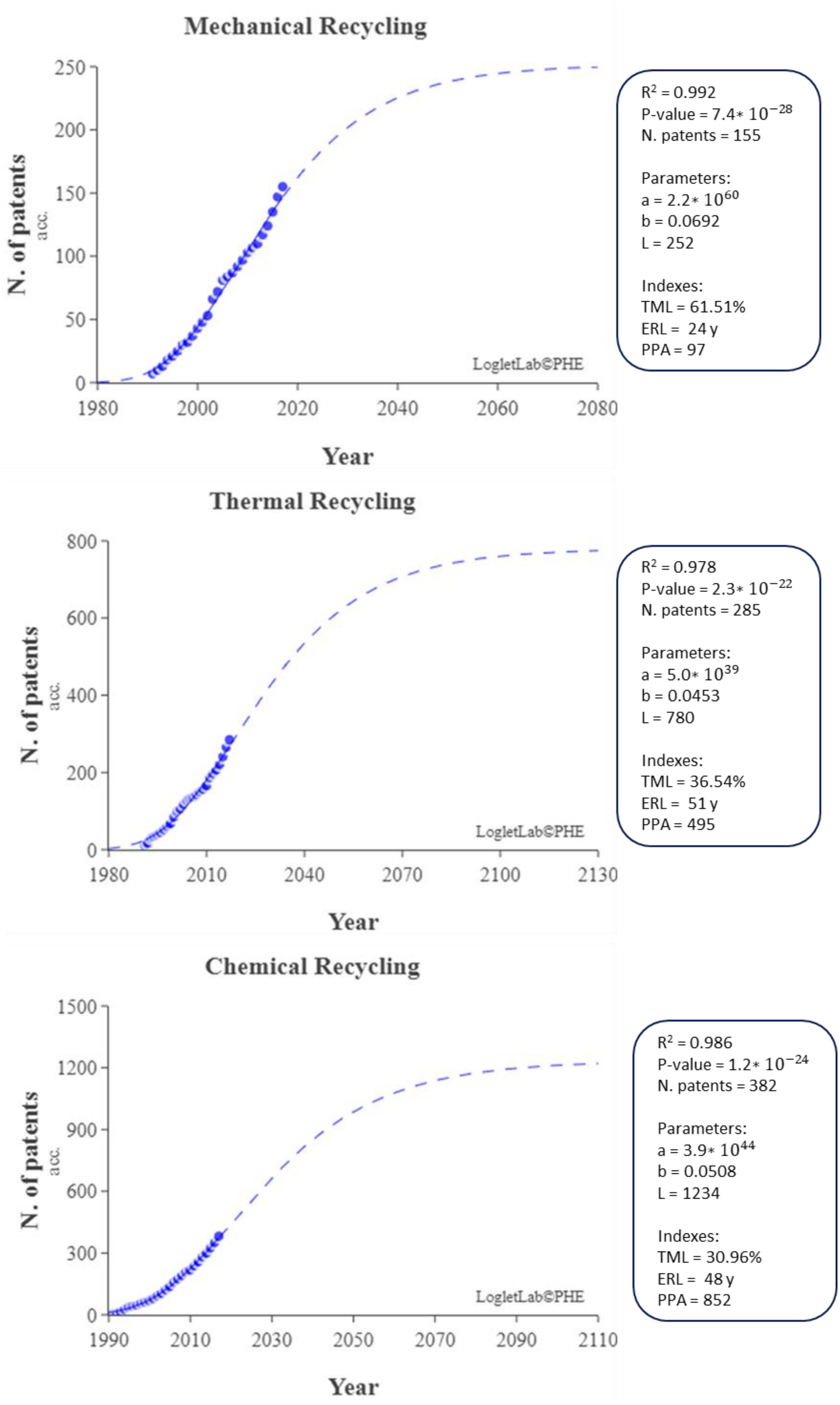


Figure 17: Historical trend of mechanical, thermal, and chemical patenting activity by first priority year (black line represents the trend of patents filed between 1991-2018, grey represents the trend of patents filed during the non-representative two-year



$R^2 = 0.992$
P-value = $7.4 * 10^{-28}$
N. patents = 155

Parameters:
a = $2.2 * 10^{60}$
b = 0.0692
L = 252

Indexes:
TML = 61.51%
ERL = 24 y
PPA = 97

$R^2 = 0.978$
P-value = $2.3 * 10^{-22}$
N. patents = 285

Parameters:
a = $5.0 * 10^{39}$
b = 0.0453
L = 780

Indexes:
TML = 36.54%
ERL = 51 y
PPA = 495

$R^2 = 0.986$
P-value = $1.2 * 10^{-24}$
N. patents = 382

Parameters:
a = $3.9 * 10^{44}$
b = 0.0508
L = 1234

Indexes:
TML = 30.96%
ERL = 48 y
PPA = 852

Figure 18: Gompertz growth curve and its parameters for mechanical, thermal, and chemical recycling technologies

Moreover, data demonstrate that patents for mechanical recycling technologies are more uniformly dispersed among industries than those for thermal and chemical recycling. This outcome can be attributed to two factors: first, mechanical recycling is a more consolidated method, and thus more widely used at an industrial level; second, it is primarily used for the recycling of GFs (Oliveux et al., 2015; Ribeiro et al., 2015), which are used in the manufacturing of a variety of applications in many industries (Sauer et al., 2018). The results also show that mechanical recycling is more interesting for industries related to industrial applications (i.e. 'Manufacture of motor vehicles, trailers and semi-trailers,' and 'Construction of buildings'). Contrariwise, patents concerning thermal and chemical recycling mostly refer to 'Manufacture of chemicals and chemical products.' This finding demonstrates that the development of chemical and thermal recycling technologies needs a high level of expertise and skills, which can be found in universities, research institutes and companies that deal with those processes and technologies.

When looking at the evolution of patents in the four main assignee groups through time, further evidence emerges. Figure 19 depicts that 'Research' grew steadily with a strong increase in the second decade of the 21st century, especially for thermal and chemical recycling. This further demonstrates that both of these recycling macro-categories are in their early stages of development and are based on considerably more sophisticated underlying principles, requiring significant research skills. In contrast, patents developed at the research level for mechanical recycling have grown much more slowly, indicating that the industrial area has made a greater contribution to the development of this technology. In terms of industries, almost all of the recycling macro-categories in "Manufacture of electrical equipment" have seen a flattening in the number of equivalent patents, indicating that this industry's technological growth is nearly complete. Furthermore, except for the 'Manufacture of motor vehicles, trailers and semi-trailers’ sector, where there is a great demand to identify solutions targeted at decreasing product and production costs, the increase in the number of equivalent patents for mechanical recycling has been marginal in recent years. Ultimately, only thermal and chemical recycling shows a growing trend in 'Manufacture of chemicals and chemical products,' because, as previously stated, these recycling macro-categories directly derive from this industry.

2.4.2.3 Citation analysis

Backward citation analysis of patents linked to FRP mechanical, thermal and chemical recycling technologies reveals possible interdependencies.

According to Table 10, where the rows correspond to the recycling macro-categories to which the citing patents belong (i.e. the patents that constitute the database) and the columns represent the recycling macro-categories to which the cited patents belong, the average number of backward citations per patent is comparable across the different recycling macro-categories. Specifically, chemical recycling has the most patents cited. Furthermore, the mechanical, thermal and chemical interdependency shares seem to be relatively similar, while the mechanical recycling macro-category has more patents referring to 'other' (90.5%) than the thermal (81.6%) and chemical (82.3%) recycling macro-categories.

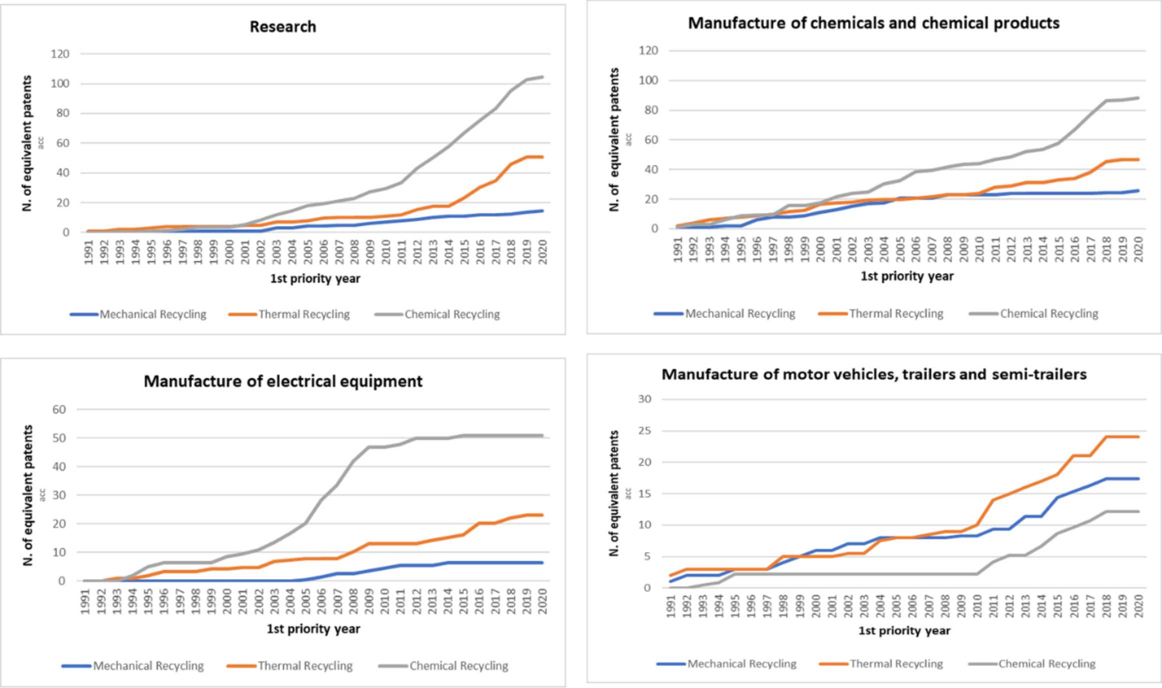


Figure 19: Cumulated patents for the main 4 assignee categories by recycling macro-category

Table 10: Distribution of backward citations per type of citation and citing patent and average number

	Mechanical	Thermal	Chemical	Other	Average number of citations per patent
Mechanical	3.3%	3.0%	3.1%	90.5%	5.9
Thermal	1.4%	11.4%	5.6%	81.6%	5.6
Chemical	0.9%	2.8%	14.0%	82.3%	6.6

This overall picture is explored in light of the development of technology citation over time in order to better grasp this distinction. Specifically, as shown in Table 11, the distribution of backward citations for mechanical recycling demonstrates a declining dependence over time on the 'other' category, despite still being high, as a result of the origin of these technologies, which have developed by borrowing technologies from other sectors.

Table 11: Distribution of backward mechanical recycling citations per type of patent cited

Year		Mechanical	Thermal	Chemical	Other
1991	1995	2.1%	1.1%	1.1%	95.8%
1996	2000	5.3%	2.7%	0.7%	91.3%
2001	2005	3.6%	3.2%	1.6%	91.6%
2006	2010	2.5%	4.1%	4.1%	89.3%
2011	2015	2.6%	2.2%	5.5%	89.7%
2016	2020	3.7%	5.2%	4.4%	86.7%

Contrariwise, as shown in Tables 12 and 13, despite the fact that both chemical and thermal recycling macro-categories heavily drew from "other" patents, this dependence significantly decreased with time in favour of a steady rise in the number of citations within the same macro-category.

Table 12: Distribution of backward thermal recycling citations per type of patent cited

Year		Mechanical	Thermal	Chemical	Other
1991	1995	0.5%	2.2%	1.1%	96.2%
1996	2000	0.0%	3.1%	2.1%	94.8%
2001	2005	0.0%	4.8%	4.3%	90.9%
2006	2010	1.9%	13.1%	6.3%	78.8%
2011	2015	1.6%	11.6%	3.3%	83.5%
2016	2020	2.9%	22.6%	13.6%	60.9%

Table 13: Distribution of backward chemical recycling citations per type of patent cited

Year		Mechanical	Thermal	Chemical	Other
1991	1995	0.4%	0.8%	1.2%	97.6%
1996	2000	0.0%	3.3%	1.6%	95.1%
2001	2005	0.4%	0.9%	11.4%	87.3%
2006	2010	1.1%	0.8%	15.6%	82.5%
2011	2015	1.0%	5.4%	15.9%	77.7%
2016	2020	1.6%	4.4%	23.0%	71.0%

According to Watts and Porter (2003), this phenomenon of convergence, which is also undoubtedly evident in the mechanical recycling macro-category, highlights a process of progressive research concentration. However, it should be noted that throughout time, there has been a rise in the quantity of patents from a certain macro-category that cite patents from other categories. As far as mechanical recycling is concerned, the citations of other categories increase in a very small way, outlining a greater orientation to internal cohesion through the consolidation of knowledge acquired from outside. Instead, chemical recycling and thermal recycling are characterised by similar self-citation patterns. Citations to the mechanical field rise in both instances, underlining its standing as a necessary prerequisite for the growth of thermal and chemical aptitude. Additionally, it is intriguing to note that there has been a progressive dependence between research in the field of thermal technologies and research in the field of chemical technologies, as evidenced by the significant increase in citations of patents from the macro-category of chemical recycling from thermal recycling patents (13.6% between 2016 and 2020). Conversely, citations of patents in the macro-category of thermal recycling from chemical recycling (4.4%) increased less considerably between 2016 and 2020, indicating stronger research independence in this field.

2.4.3 Hints about potential future trends

Some conclusions concerning the potential development of FRP recycling technologies in the future can be taken from the findings revealed in the previous sections of sub-section 2.4. First, given the lower degree of technological maturity of the thermal and chemical recycling macro-categories, it is reasonable to anticipate that the number of patents will increase gradually and continuously in the near future. Overall, it is reasonable to predict that patent growth will accelerate as a result of increased social pressure, legislative restrictions and market sensitivity to the environmental sustainability issue (Dyllick and Muff, 2016). With regard to mechanical recycling, given that the technology has reached a point where the quality of the recycled raw material is not very high (Oliveux et al., 2015; Pickering, 2006), it is reasonable to anticipate that, except for disruptive innovations, research in this area will have to focus primarily on production system reliability. It is accepted that after a certain technological limit is achieved, the focus of the research turns to other areas that are more closely tied to the production process and its reliability (Mann, 2002).

Contrariwise, future research in thermal and chemical recycling macro-categories should continue to concentrate on reducing the inefficiency of current technologies (Mann, 2002), as the time series analysis suggests there is still significant room for improvement. In addition, industry-oriented research in mechanical recycling should take precedence over basic research in the future, with a particular emphasis on industries where the need is to create the most favourable conditions for minimizing product and production costs rather than to have high-quality products for structural applications. The automotive sector is a prime example of this (Sauer, 2019), which is characterised by a growing interest in lightening the weight of cars and enhancing driving dynamics by employing less expensive and more environmentally friendly materials in order to lessen environmental effect and comply to rules. In such a context, it is possible to assert that, while being marginal, innovations for mechanical recycling will likely originate from this industry, where the importance to use FRP composite materials' light weight even for non-structural components emerges as essential. In contrast, research on thermal and chemical recycling will still be adopted to recover raw materials that might be appropriate for making structural goods. Additionally, research in these areas will continue to be produced by academic institutions and research institutes, where fundamental research is more ingrained, as well as by those industries that have specialized knowledge of technology and process, at least in the near future. These sorts of recycling unquestionably need considerable upfront investments in terms of expertise and material and financial resources that cannot be afforded by all businesses, indicating that incumbent firms would likely do research in this area. Nevertheless, the study focus will gradually change from the standpoint of the transformation process to that of the transformation system as expertise in this field develops, encouraging spread into other industries generally served by mechanical recycling technology. It is also realistic to anticipate that mechanical recycling research will become more self-centred. In fact, unless a disruptive technology is invented, the objective of prospective new patents will be to improve recycling process reliability. Once consolidated, research will be limited to marginal activities since it has reached its natural limit. In contrast, as it is still in the expansion phase, research on the mechanical technologies will continue to have a higher level of cohesiveness in the next years. However, this difference is anticipated to narrow with time as research consolidates. Eventually, it can be asserted that research entropy, i.e. irregularly drawing from different macro-categories over time (Miranda et al., 2019), will significantly increase, implying that studies on chemical and thermal recycling will tend to converge into a single field of interest. This is supported by an increasing level of cross-citations between chemical and thermal macro-categories.

2.4.4 Limitations and future research paths

The patent analysis performed has some limitations. First, it was conducted under the assumption that patents are a trustworthy and valid source of technological information. Nevertheless, not all innovations may be patented as the trend in patenting might differ from nation to nation and from industry to industry and because patenting is not always the greatest way to safeguard an innovation (*OECD Pat. Stat. Man.*, 2009). Second, there are some issues with the method utilized to identify pertinent patents. On the one hand, uneven terminology among businesses, inventors and patent grantors is the fundamental drawback of the keyword-based method (Karvonen et al., 2016). On the other side, using numerous IPC codes might both result in a large number of patents that are not relevant and make it challenging to research a certain area of interest (Pilkington et al., 2002). These limitations were exceeded as much as possible by performing the manual filtering of the retrieved patents. To improve the supply chain for the composites material industry and the synergy between recycling technologies, it is recommended that future research focus on potential industrial outlet sectors for recycled FRP composite materials as well as the accomplishment and management of potential industrial symbiosis. Furthermore, the analysis performed could be expanded through the application of additional patent analysis methodologies to examine the viewpoint of key technologies and changes to a specific technical sector.

2.5 Conclusions

A bibliometric analysis and a patent analysis have been carried out to provide a comprehensive and holistic overview of the existing FRP recycling technologies, as well as to assess their actual maturity level. In doing so, it has also been possible to identify literature hotspots and gaps, as well as future research directions.

Since it turned out that plenty of studies exist on the research and development of recycling technologies to improve the quality of recycled fibre, but scant attention has been paid to the production methods of FRP composites using recycled fibres for structural applications, within this Ph.D. project, it was decided to focus on this last aspect. This choice is also in line with the principles of the circular economy emphasising the use of re-use practices over recycling practices (Ellen MacArthur Foundation, 2013).

Both analyses highlighted that the interest in the topic has been gaining momentum over the last decades likely owing to the increased market knowledge of the sustainability issue and the pressure from the authorities (Colombo et al., 2022, 2021). However, scientific interest has been, and still is, different for each of the recycling macro-categories. In detail, the bibliometric analysis showed that mechanical recycling has been less studied than other recycling technologies and scientific interest has declined over time. Currently, it is mainly used to reduce the size of composite materials waste whose fibre are supposed to be recovered through more complex recycling technologies, since they do not significantly impact on the mechanical properties of the recovered fibres. Therefore, nowadays, mechanical recycling is considered a pre-recycling process. These results are fully consistent with the fact that such a technology is already in its maturity stage, as emerged by means of the patent analysis. Instead, thermal recycling and chemical recycling are the most compelling macro-categories. According to the patent analysis, they are in the growth stage of their life cycle and have a significant potential of growth; therefore, they seem to be the most promising recycling technologies. Such an outcome is corroborated by the bibliometric analysis which showed that the most common research methodology adopted, so far, is the experimental study. Moreover, industries with specialized technology and process knowledge, as well as universities and research institutions, where basic research is more ingrained, will continue to develop patents in these categories in the future. The same may be applied, at least in the next few years, for scientific research since there is ample room for improvement especially considering the increasing attention to the sustainability issue.

Lastly, to the best of the researcher’s knowledge, both analyses represent the first attempts to exploit the rigorous methodologies used to achieve the goal set. The findings may provide invaluable advice for academics, non-academic researchers and practitioners. The bibliometric analysis may help academics identify the main research streams and gaps to study in future investigations, while it may help practitioners gain a full understanding of current trends in FRP recycling technologies. These trends are particularly stressed by the patent analysis whose results may be useful for businesses to understand where to target future technological investments. Obviously, such an investigation is also essential for researchers. On the one side, academic researchers can discover new areas of development and figure out where to direct research efforts. On the other side, non-academic researchers can pinpoint where to focus their efforts toward applied research (i.e. to turn scientific study results into real-world applications).

CHAPTER 3

The innovative spinning process

3.1 Introduction

Utilizing non-woven fabrics and yarns in processes to produce value-added products is crucial in light of the circular economy (Pakdel et al., 2020). Herein, scholars agree that the spinning process technology has potential to handle recycle CFs to produce CFRPs for structural applications. Although the benefits are well known, the topic at hand has not yet received much attention in the scientific literature with the development of few studies on this issue. While Akonda et al. (2012) and Akonda et al. (2014) used wrap spinning, Hengstermann et al. (2016b) and Hengstermann et al. (2017) used a roving frame to produce hybrid yarns. Specifically, Hengstermann et al. (2016b) developed yarns consisting of polyamide 6 and virgin CF that was cut to replicate recycled CFs from manufacturing scrap at lengths of 40 and 60 mm. In particular, the authors discovered that the length and ultimate concentration of CFs within the yarn impact its physical and mechanical properties. The tensile strength of the generated unidirectional composites, which was discovered to be between 771 and 838 MPa, is also influenced by these parameters. Additionally, to manufacture hybrid yarns, Akonda et al. (2012) utilised recycled CFs with a length of around 50 mm derived from thermal recycling fibres and PP fibres with blending ratios of 30–70% and 50–50% by weight, respectively. The investigation of the mechanical characteristics revealed that the mixing ratio is essential since the tensile strength increases with increasing fibre amount and length. Furthermore, Hasan et al. (2018) produced friction spun core-sheath hybrid yarns using polyamide 6 and virgin CFs cut to obtain 60 mm length. They discovered that the core to sheath ratio of the hybrid yarns and the air suction required to produce them have an impact on the mechanical properties of unidirectional thermoplastic composites.

All in all, the findings from earlier study are encouraging despite some technological limitations. However, the production techniques exploited until now do not guarantee the manufacture of high-quality yarns. Therefore, further efforts should be made to find out the most effective spinning process for making value-added use of waste and contributing definitively to closing the loop.

In summary, to date, an innovative spinning process involving preparation, carding and drawing has been developed in the literature for CFs recycled from manufacturing scraps, using virgin fibres cut to simulate them. Moreover, ring spinning has not yet been explored for the production of hybrid yarns suitable for CFRPs for structural applications although it allows for the production of hybrid yarns of a quality level that makes them excellent for the production of fabrics that may be used for a variety of final uses as well as for the processing of fibre mixes in addition to the majority of natural and synthetic fibres (Lawrence, 2003). In greater detail, blends of CF with polymer fibres were used, consistently with existing literature, as they allow the production of slivers and subsequent finished products with superior mechanical properties. Additionally, a 100% CF input would increase the percentage of fibres sticking to the card clothing and its electrical conductivity, negatively impacting yarn production.

Considering the above, the researcher decided to propose an innovative spinning process composed of all the steps of the traditional spinning process where the spinning phase leverages the ring spinning technology. Furthermore, the outcomes presented are particularly reliable since the raw material used comes from actual manufacturing scraps.

In such a context, this Chapter aims to present the innovative spinning process developed at laboratory level in order to manufacture ring-spun hybrid yarns suitable for the production of reinforcement capable of improving the properties of the final CFRPs as well as the properties of the different ring-spun hybrid yarns produced. The remainder of this Chapter is organised as follows. First, a brief overview of the traditional spinning process is provided. Second, the Chapter presents the innovative spinning process developed and its technical feasibility. Then, the physical-thermal and mechanical analysis of the ring-spun hybrid yarns produced is shown. Furthermore, the methods used and the outcomes obtained are explained in detail.

3.2 The traditional spinning process

One of the oldest manufacturing techniques is the mechanical spinning of staple fibres into yarns (Kadolph, 2010). Overall, the fundamentals of spinning have not changed since the invention of yarn. Clearly, improvements in engineering and technology have sped up spinning and increased the amount of yarn manufactured. For instance, after the industrial revolution, when power machines replaced manual labour and enabled mass production, several innovations for enhancing spinning emerged (Kadolph, 2010). In detail, specific machineries were developed for each distinct step of the process. Nowadays, further improvements in machinery and process parameters are still simplifying and automatizing the procedure, enhancing yarn quality.

Traditional yarns are typically produced using a multi-step process that can be divided into four main stages, namely preparing, carding, drawing and spinning (Lawrence, 2003). Each step is essential and has a specific purpose that makes it central to the whole process, which is sketched in Figure 20. It is worth noting that sometimes, according to the type of fibre used and the final application of the yarn produced, certain steps (e.g. combing) may be added. The individual steps are discussed in more detail below.

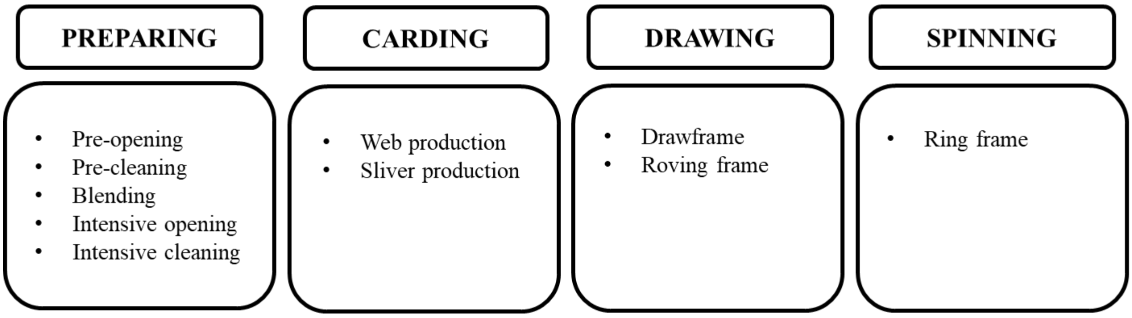


Figure 20: Main steps of a traditional spinning process

3.2.2 Preparing

Fibres, especially natural ones such as cotton and wool, require preliminary cleaning, opening and blending before they can undergo the multiple stages of spinning. Such operations are carried out to achieve greater uniformity in the quality of the final yarn produced (Kadolph, 2010). As each natural textile fibre intrinsically possesses its own length and raw state of presentation, there is no univocal preparation process (Bona et al., 1981). Several machines are often set up in a sequential order to clean and blend the fibre mass (Lawrence, 2003). All in all, the ultimate output of this phase is a *proper fibre blend* suitable for feeding the next phase.

3.2.3 Carding

Carding plays a fundamental role within the whole spinning process (Lawrence, 2003). Indeed, it is directly responsible for the final properties of the yarn. The main purpose of this step is to separate the fibres, thus completing the opening and cleaning begun in the previous step (Bona et al., 1981). Concurrently, thanks to the inherent architecture of the machine, both length-based fibre selection and particularly efficient blending take place (Bona et al., 1981; Lawrence, 2003). By parallelizing the fibres, the carding constitutes a thin web from which a delicate, incredibly frail rope of fibres known as *carded sliver* is formed through the coiler. A sliver is a fibre strand that resembles a rope (Kadolph, 2010). The carding operation occurs through the carding machine which is composed of a rotating cylinder and some *card clothing* with fine, short wire teeth (Kadolph, 2010).

3.2.4 Drawing

Drawing turns multiple carded or combed slivers into a single *drawn sliver* and significantly enhances the parallelism of the fibres (Kadolph, 2010; Lawrence, 2003). Such a doubling operation increases the uniformity of yarn. The machineries to perform the drawing phase are known as *frames*. The drawframe consists of a set of rollers that are coupled together to form a drafting zone. Generally, more than one-zone drafting

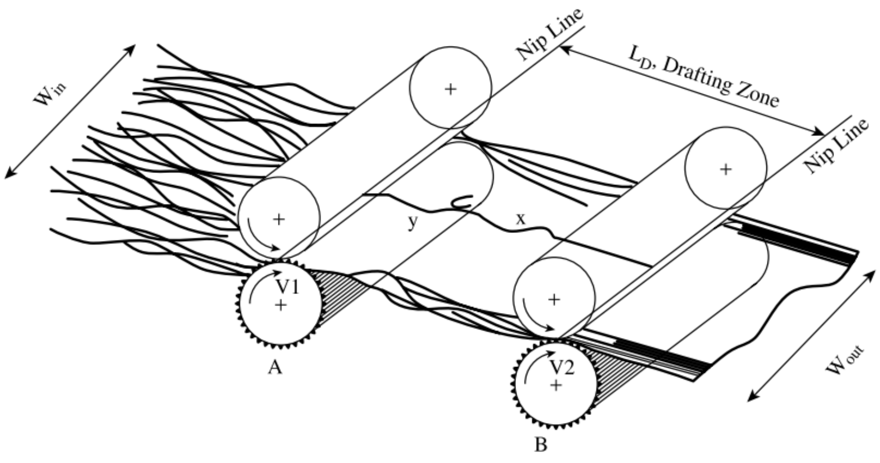


Figure 21: One-zone drafting arrangement (from Lawrence (2003))

arrangement (Figure 21) is used in order to control how material properties affect drawing inconsistencies. Furthermore, each set rolling more quickly than the previous one. The drafting zone comprises a *break draft* zone that helps to reduce the negative effects of sliver extension in the following higher draft zone, called *main draft* (Lawrence, 2003). The outcome of this phase is a thinner sliver, since more slivers (usually from 2 to 8) are combined (Kadolph, 2010). Such a result is then turned into a *roving*, usually by means of a roving frame (but other methods exist such as *flyer frame* and *speed frame*). In detail, a roving is a soft continuous fibrous strand characterised by discrete cohesiveness due to slight twist (Lawrence, 2003).

3.2.5 Spinning

As regards the spinning phase, a large variety of spinning systems exist, but not all of them are widely exploited in commerce. The most popular one within textile companies is *ring or conventional spinning* (Xu et al., 2011), which holds about 90% of the global spinning machine market (Lawrence, 2003). The other systems (e.g. open-end spinning, wrap spinning etc.) are defined as *unconventional spinning processes*. Since its invention in the 19th century, ring spinning dominated the market owing to different factors such as the high quality of yarns obtained, the large range of applicability of raw materials and the versatility of yarns counts it allows to produce (Goyal and Nayak, 2019; Yin et al., 2021). The ring or conventional spinning (Figure 22) uses roller drafting to reduce the mass of the fibres and the action of a guide, known as a traveller, freely circling around a ring to introduce twist while also winding the created yarn onto a bobbin (Lawrence, 2003). Its purpose is "to (1) clean and make parallel staple fibres, (2) draw them out into a fine strand and (3) twist them to keep them together and give them strength" (Kadolph, 2010). Indeed, the yarns produced are smooth, of good-quality and homogeneous; therefore, they are suitable for the vast majority of end applications (Lawrence, 2003). Furthermore, their tensile strength is the highest when compared to yarn made with other spinning technologies (Yin, 2021). Among others, it is the most versatile and cheapest spinning process and its maintenance cost is the lowest (Kadolph, 2010; Yin, 2021).

Nevertheless, it has also some drawbacks (Lawrence, 2003). The most prominent concerns the speed of the process. Indeed, ring spinning is slow (rates range from 25,000 to 30,000 rpm) rather than open-end spinning (which can reach 150,000 rpm). This fact rides on the frictional contact of ring and traveller as well as yarn tension. To ensure a high production speed, the yarn package has to be downsized. However, this leads to frequent stops for doffing and the need to rewind yarns to fabricate larger size packages. Furthermore, although it is well-recognised that the spinning process involving various mechanical processes is energy-intensive (Yin et al., 2021), ring spinning emerges as the most polluting due to the higher number of machinery required for the process (Goyal and Nayak, 2019).

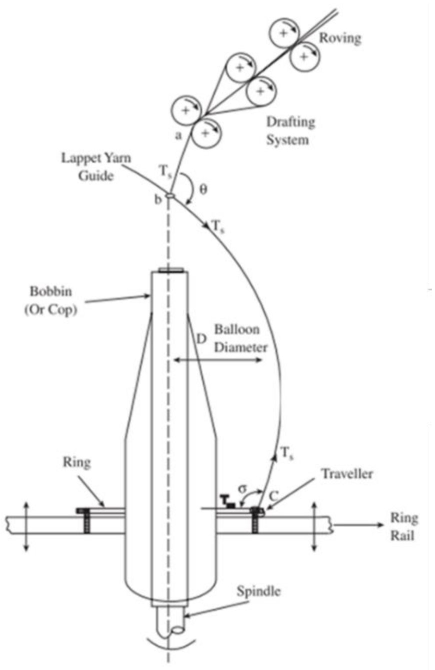


Figure 22: Ring spinning system (from Lawrence (2003))

3.3 The proposed innovative spinning process

It is common knowledge that a traditional spinning process may not handle CFs due to their brittleness, their sensitivity to shear stresses and their crimp-free nature. To achieve the goal set, an innovative spinning process has been developed. Of course, it involves the same phases of the traditional spinning process, as depicted in Figure 23.

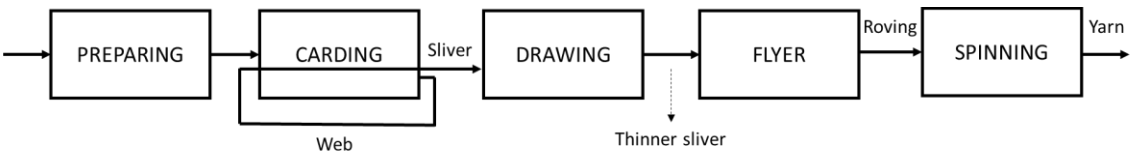


Figure 23: Phases of the proposed innovative spinning process

Nevertheless, some adjustments have been made in order to suit the characteristics of CFs. In detail, it encompasses a manual mixing phase (i.e. preparing) (Figure 24) in order to open the different fibres, a double carding (one passage for the production of the carded web, and one passage for the production of the sliver) shown in Figure 25, a drawing to fabricate a thinner sliver followed by the flyer to manufacture the roving (Figure 26 and Figure 27) and finally the ring spinning to produce the yarn (Figure 29).

Figure 24: Manual mixing phase

Figure 25: Carding phase of the innovative spinning process

The actual innovation of the proposed spinning process with respect to the handling of recycled CFs lies in the use of the ring-spinning technology as well as in modifications to specific machinery and process parameters. To arrive at the proposed changes, a challenging trial-and-error exercise was conducted, involving numerous attempts. At first, the focus was on machinery and only subsequently on process parameters.

As the carding phase plays a pivotal role within the entire spinning process, efforts have focused on this machinery. In detail, a low dens card clothing was applied to assure an extremely delicate carding process on recycled CFs. Then, an inverter was installed on the carding machine to better control and regulate the speed of the doffer and, accordingly, avoid web breakages. At process parameter level, the speed of the feed roller of the carding machine was modified. Finally, the gauge of the drafting zone within the drawing phase was revised (details are shown in Figure 29).

Figure 26: Drawing phase of the innovative spinning process

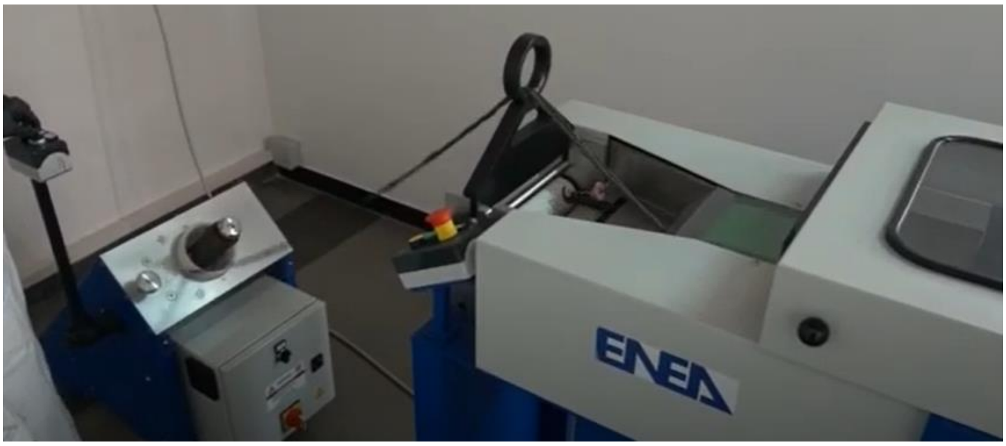


Figure 27: Flyer machine used for the production of the roving

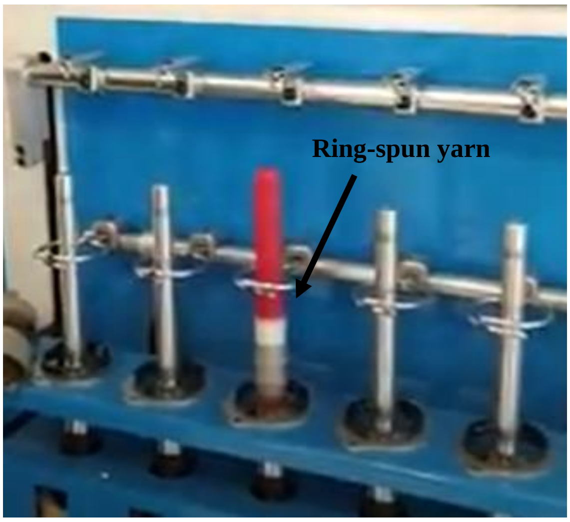


Figure 29: Ring spinning machine adopted with ring bobbin production

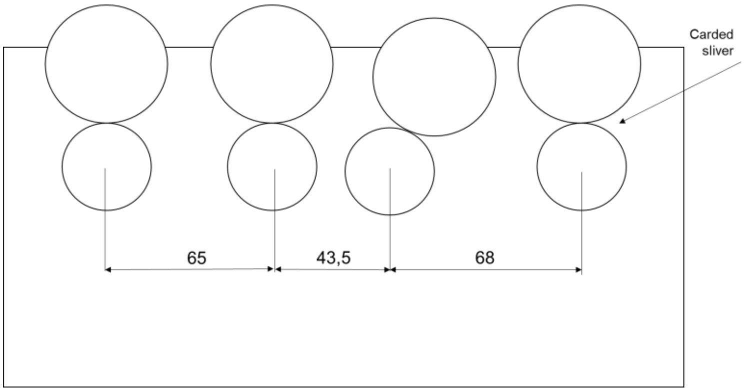


Figure 28: Clearance between the different drafting groups (values in millimetres)

3.4 Materials and methods

3.4.1 Materials

An Italian company with expertise in the production of fabrics made up of CF provided the recycled CFs used in this investigation. Specifically, CFs recovered from manufacturing scraps were adopted since they constitute the primary source of CFRPs waste, accounting for approximately 40% of the total (Pickering, 2006; Pimenta and Pinho, 2011). In greater detail, rCF of dry fabrics (unidirectional or weaving), already opened by the supplier through a mechanical process to untangle the weft and warp threads or unidirectional wefts, was used. Thus, no pre-processing treatment was performed. Additionally, from a technical standpoint, they resemble virgin CFs in terms of its properties, making their processing easier (Hasan et al., 2018). Recycled CFs employed in this study are shown in Figure 30.

Figure 30: Recycled CFs used for the investigation

The average length of the recycled CFs declared by the provider is 62 ± 5 mm. Polyester and polyamide staple fibres of the same length as the CF were used as carriers to facilitate the process. These thermoplastic fibres were chosen for two main reasons. First, they are characterised by excellent fibre-to-fibre cohesion and entanglement (Akonda et al., 2014; Hengstermann et al., 2016b), thus improving the mechanical properties of the carded web. Second, they could be used as matrix components for the production of thermoplastic CFRPs (Hasan et al., 2018; Hengstermann et al., 2016b). Furthermore, the use of white polyester and polyamide staple fibres together with black CFs made it possible to visually evaluate certain characteristics of the semi-finished products, such as the presence of holes and the orientation of recycled CFs. Finally, it is worth mentioning that to limit researcher bias as much as possible, the fibres used in all tests were supplied by the same provider.

Normally, there is a not a technical datasheet for recycled CF commercially available, since it frequently does not have a fixed length. As a result, the researcher identified a few distinct characteristics, including diameter distribution, surface quality and tensile strength and modulus by means of specific laboratory tests described in sub-section 3.4.2. The virgin fibres used for comparison purposes have tensile strengths of 4,900 MPa, tensile modulus of 230 GPa, a density of 1.8 g/cm^3 and an average diameter of 7 µm. They are commercially available from Torayca under the name T700S.

3.4.2 Methods

This sub-section reports first the different laboratory tests carried out to define the diameter distribution, surface quality and tensile strength of the fibres utilised. Second, the technical feasibility study of the proposed innovative spinning process. In doing so, the most appropriate range of blending ratios by weight to obtain ring-spun hybrid yarns with acceptable properties for the production of CFRPs for structural applications was identified.

3.4.2.1 Characterisation of recycled CF

The statistical distribution of the diameter was evaluated using a statistical sample of 30 measurements. A LEICA DVM6 (Figure 32) optical microscope was used to perform these measurements. Instead, a Zeiss LEO 1530 SEM equipment scanning electron microscope was used to conduct a morphological analysis of the surface of the fibres. Single-filament tensile tests were carried out using a Zwich Roell 1 K dynamometer with a 5N loading cell (Figure 31). Samples were set up by bonding a single filament to cardboard cut to gauge lengths of 10, 20 and 40 millimetres, which was subsequently clamped between the dynamometer's tools. In accordance with ASTM 3379, tensile tests

Figure 32: The LEICA DVM6 optical microscope used to obtain diameter measurements

Figure 31: Zwich Roell 1 K dynamometer with a sample entered between the grips

were conducted utilizing a pre-load of 0,002N and applying the load at a constant crosshead rate of 1 mm/min. In detail, 30 repetitions for each gauge length were executed. Tensile strength and elastic modulus are both computed using the previously determined nominal value of the diameter of fibres.

3.4.2.2 Technical feasibility study

In order to understand whether the innovative spinning process is able to handle recycled CF from manufacturing scraps and, accordingly, identify the range of recycled CF allowing a proper balance between tensile strength and real amount of recycled CF for the ring-spun hybrid yarns, a technical feasibility study was carried out.

To this end, 25 grams of fibre mass consisting of recycled CFs and polyester or polyamide in different blending ratios by weight were utilised and a number of control points were placed along the innovative spinning process. Specifically, in order of execution, these are two visual checks and two quantitative checks (Figure 33).

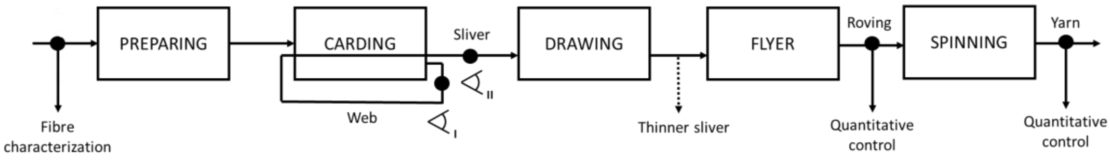


Figure 33: Control points along the proposed innovative spinning process

The adopted control process was pass-fail. As a result, if a blend failed an initial visual inspection, no further checks were performed on it and the blend was directly discarded. Below, the three different types of control are explained in detail.

Visual controls

Two experts performed two subsequent visual inspections. In accordance with Hengstermann et al. (2016b), two consecutive visual controls were carried out to analyse the orientation of the CF in the semi-finished products. Specifically, the web was subjected to the first inspection after it had passed through the carding machine once, while the carded sliver was subjected to the second inspection when it came out of the coiler. During each inspection, the surface of the semi-finished product was examined for holes or defects. Subsequently, the homogeneity of the individual outputs and the direction of the CFs were evaluated thanks to the difference in colour between the two fibres used.

Quantitative controls

Finally, two quantitative controls were carried out. First, a thermal investigation, namely a pyrolysis under nitrogen, was used to determine how many CFs were still present in the roving, the last semi-finished product of the spinning process. Specifically, to extract the thermoplastic fibres without affecting the CF, a specimen weighing about 150 mg per roving was prepared. The various specimens were placed in crucibles after being precisely weighed to the fifth decimal figure. They were then placed inside the muffle, which had been preheated to 200°C with nitrogen (Figure 344). To make sure that the thermoplastic fibre in the roving is fully decomposed, a crucible containing only the thermoplastic fibre was also placed inside the muffle. A ramp of around 20°C/min was used to reach a temperature of 500°C. After approximately 45 minutes of standstill, cooling (speed below 2°C/min) was established. The nitrogen flow was stopped at about 300°C and post-oxidation was performed to ensure a clean surface for CFs. In detail, the samples were stalled for 15 minutes at a temperature of roughly 450°C. They were finally cooled in air. By weighing the residual mass of recycled CF, the percentage composition of recycled CF in the finished roving was calculated.

Figure 34: Crucibles within the muffle

Second, various laboratory tests were carried out to evaluate the thermal-physical and mechanical properties of the fabricated hybrid yarns. These tests are reported in the following.

- **Count**: Three samples were taken into consideration while determining the mean count of the hybrid yarns. For each sample, the count was determined by means of the well-known direct *dtex* counting formula (Eq. (2)):

$$10000 \cdot \frac{P\ (g)}{L\ (m)} \tag{2}$$

where P is the weight in grams of the skein of yarn with a 5 m length.

- **Tensile properties**: Tensile testing of the fabricated yarns were performed using an MTS ALLIANCE RT50 2 kN dynamometer (Figure 355). In accordance with ISO 3341, the sample length was set up to 500 mm for each hybrid yarn. In particular, flat clamps were used to complete 10 repetitions at a constant crosshead speed of 200 mm/min. Using the previously determined diameter values for each

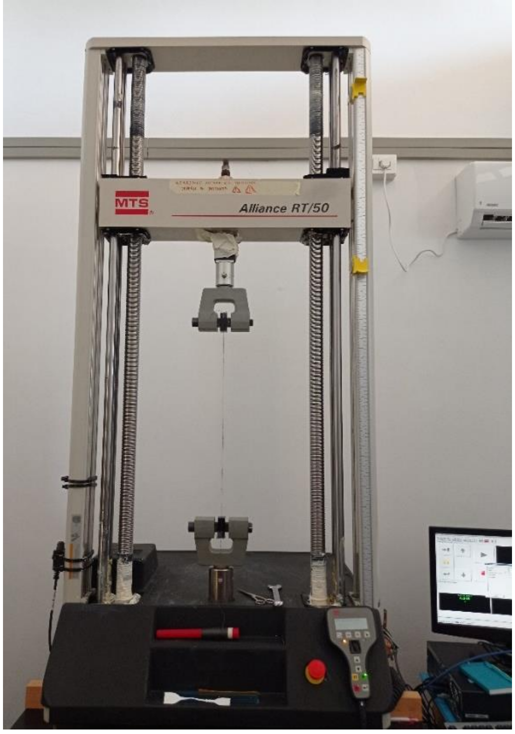


Figure 35: MTS Alliance RT50 2kN dynamometer used with a sample inserted between the clamps

hybrid yarn, Testworks®4 software was used to compute the elastic modulus and tenacity.

- **Thermogravimetric analysis and Differential scanning calorimetry**: Through the TA instruments SDT Q600 (Figure 366), a simultaneous thermal analyser, a thermogravimetric analysis (TGA) and a differential scanning calorimetry (DSC) were simultaneously performed. In order to precisely measure the quantity of recycled CFs present in the hybrid yarns, TGA was carried out. Instead, DSC was used to analyse the melting behaviour of the thermoplastic fibres. In detail, the yarn samples that were put into the machinery weigh about 10 mg. Starting from room temperature (25°C), a linear heating rate of the thermal cycle was set at 10°C/min until 800°C was reached. Each type of hybrid yarn was subjected to three measurements. To ensure an inert atmosphere, all samples were examined under nitrogen.

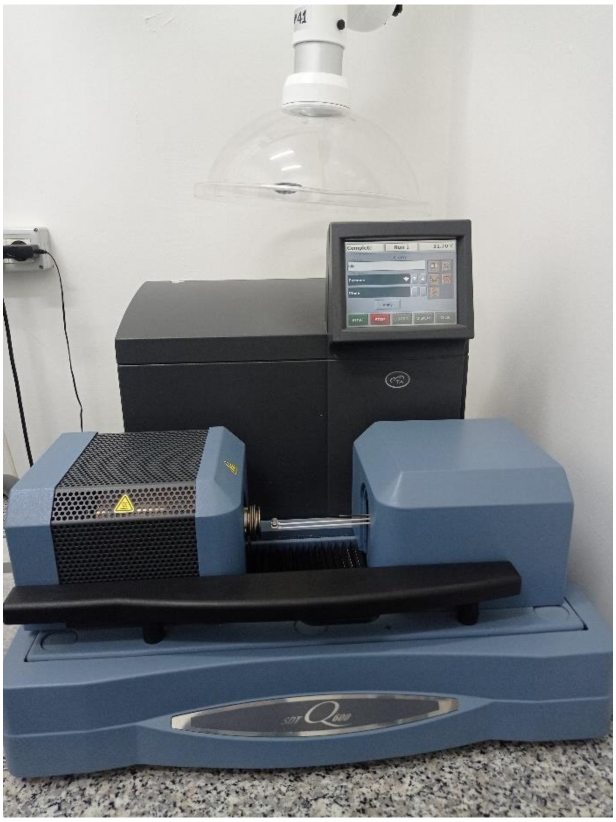


Figure 36: TA instruments SDT Q600 used for TGA and DSC

Hybrid yarns were produced through the innovative spinning process described in Section 3.3, by using 25 grams of fibre mass consisting of recycled CFs and polyester or polyamide in different blending ratios by weight. The complete spinning plan is detailed in Table 14.

Table 14: Complete spinning plan for the different mixing ratios (PL: polyester; PA: polyamide, 3 and 5 are the number of draw frame doublings)

			Carding	**Drawing + Flyer**		**Spinning**
Sample	**CF length [mm]**	**% rCF**	**Carder sliver [ktex]**	**Doubling**	**Roving [ktex]**	**Yarn [dtex]**
rCF-PL3$_{50}$	62 ± 5	50%	1.72	3	1.16	580
rCF-PL5$_{50}$	62 ± 5	50%	1.19	5	1.34	447
rCF-PA3$_{50}$	62 ± 5	50%	1.90	3	1.28	640
rCF-PA5$_{50}$	62 ± 5	50%	1.82	5	2.04	680
rCF-PL3$_{60}$	62 ± 5	60%	1.87	3	1.26	630
rCF-PL5$_{60}$	62 ± 5	60%	1.13	5	1.27	424
rCF-PA3$_{60}$	62 ± 5	60%	2.02	3	1.36	681
rCF-PA5$_{60}$	62 ± 5	60%	1.87	5	2.09	698
rCF-PL3$_{70}$	62 ± 5	70%	1.49	3	1.00	501
rCF-PL5$_{70}$	62 ± 5	70%	1.33	5	1.50	500
rCF-PA3$_{70}$	62 ± 5	70%	1.50	3	1.01	507
rCF-PA5$_{70}$	62 ± 5	70%	1.86	5	2.09	695

Plan of experiments

The experimental campaign was conducted on the base of a 3 x 2 x 2 full factorial design of experiments (DoE) by varying the amount of recycled CFs (rCF) on three levels, while the type of thermoplastic fibre (Thf) and the number of draw frame doubling (Ndf) on two levels, namely polyester or polyamide and 3 or 5, respectively. Three repetitions were considered for each combination of the parameters. Table 15 shows the 3 x 2 x 2 factorial plan displaying the combination of parameters.

In detail, as regards rCF parameter, 50%, 60% and 70% were selected considering the range found thanks to the technical feasibility study. The Ndf parameter was set to 3 or 5 to assess a potential improvement in yarn properties as the number of doubling and, consequently, fibre parallelisation increases. Besides, these are values usually used at an industrial level. Finally, as regards Thf, polyester and polyamide were selected since they act as carriers for CF and are widely used in the textile industry. They are summarised in Table 16.

The analysis of variance (ANOVA) was adopted to verify their potential influence on the remaining amount of recycled CF of the produced yarns. To this end, a confidence interval equal to 95% was considered.

Table 15: Design of experiments - Parameters combinations

Samples	rCF	ThF	Ndf
1-13-25	50	PA	3
2-14-26	60	PA	3
3-15-27	70	PA	3
4-16-28	50	PL	3
5-17-29	60	PL	3
6-18-30	70	PL	3
7-19-31	50	PA	5
8-20-32	60	PA	5
9-21-33	70	PA	5
10-22-34	50	PL	5
11-23-35	60	PL	5
12-24-36	70	PL	5

Table 16: Parameters and related levels

Parameters	Abbreviation	Low level	Medium level	High level
Amount of recycled CF [%]	rCF	50	60	70
Thermoplastic fibre	Thf	PA	-	PL
Number of draw frame doubling	Ndf	3	-	5

3.5 Results and discussion

This section reports the outcomes obtained from the characterisation of the recycled CF and the different control points described in sub-section 3.4.2.

Characterisation of recycled CF

A SEM micrograph of the CFs used in this investigation is shown in Figure 387. Overall, the surface appears smooth and free of carbonaceous deposits. Moreover, the statistical distribution of the diameter analysed using a statistical sample of 30 measurements is depicted in Figure 378. The average diameter is 7.27 ± 0.72 µm.

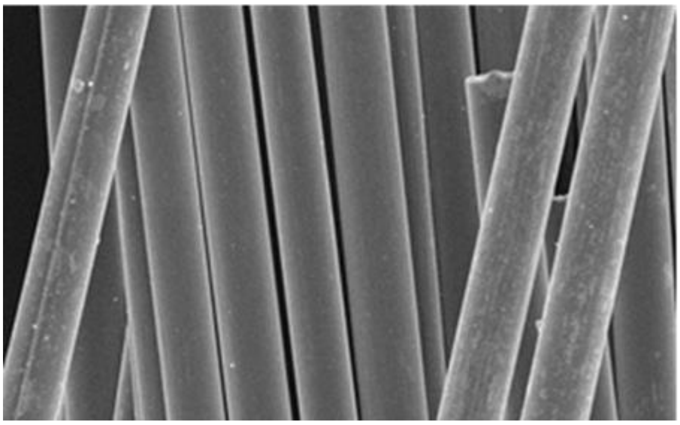


Figure 38: Surface of recycled CFs used

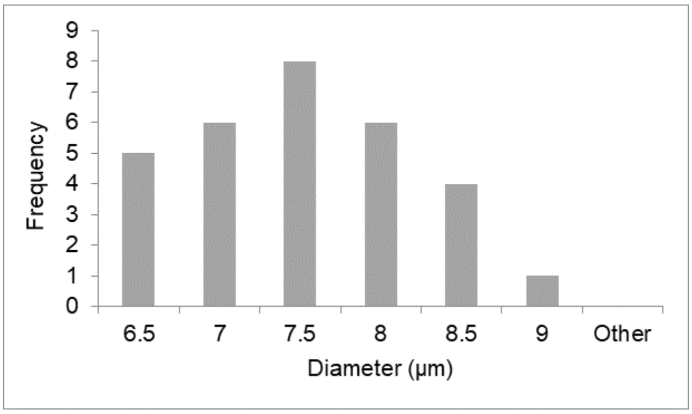


Figure 37: Statistical distribution of the diameter of recycled CFs evaluated with a sample of 30 measurements

The Weibull statistic, which is typically used to explain the strength distribution of single fibres under the hypothesis that defects exist, was used to interpret the strength data (Fu and Lauke, 1998; Garoushi et al., 2006). Weibull distribution consists of two parameters, the scale parameter and the shape parameter. The former represents the stress corresponding to a failure probability of 63.2% of the specimens, while the latter represents the slope of the distribution and it is also known as the Weibull modulus.

By linearizing the cumulative distribution function as shown in Eq. (3), the scale (σ_0) and shape (β) parameters, whose values are shown in Table 17, were identified.

$$\ln\left(\ln\left(\frac{1}{1-F}\right)\right) = \beta\ln(\sigma) - \beta\ln(\sigma_0) + \ln(AL_f) \tag{3}$$

where L_f is the gauge length of a single fibre during the test, A is the fibre cross section, σ is the single fibre's tensile strength and F is the probability of failure.

Table 17: Weibull statistics parameters for the recycled CF used

Single fibre length	σ_0 (MPa)	β
rCF (10 mm)	3802	5.41
rCF (20 mm)	3601	4.08
rCF (40 mm)	3368	4.70

In particular, F was calculated using experimental tensile strength data using a median-order approach (Davidge, 1978), following Eq. (4):

$$F = \frac{i - 0.3}{N + 0.4} \tag{4}$$

where N is the total number of samples and i is the i-th stress value. Figure 40 illustrates a plot example with its linear-fitting method.

Then, taking into account a specified breaking probability and inverting Eq. (3), the tensile strength was found by varying the fibre length (L), as the former rides on the latter. Eq. (3) thus becomes Eq. (5).

$$\sigma = \sigma_0\left(\frac{L_f}{L}\right)^{1/\beta} \tag{5}$$

Figure **39**40 displays the graphs derived from Eq. (5) using the values from Table 18 and experimental data collected. Overall, it can be concluded that the Weibull model accurately represents the experimental data.

To summarise, the properties of rCF, virgin polyester fibre and virgin polyamide fibre are reported in Table 18.

Lastly, the load-extension curves from the single fibre tensile test of 30 fibres per each gauge length (10, 20 and 40 mm) are shown in Figure 41. Overall, the results show that as the length increases, the average tensile strength values decrease. This may be traced back to the fact that as the length of the fibre increases, so does the surface area considered and, therefore, the probability of finding defects.

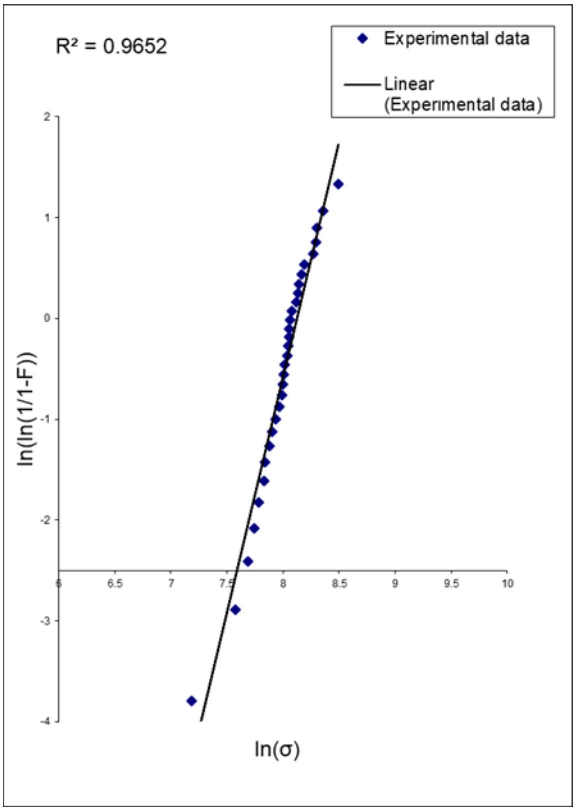


Figure 40: Weibull linear-fitting plot for recycled CFs of 40 mm gauge length

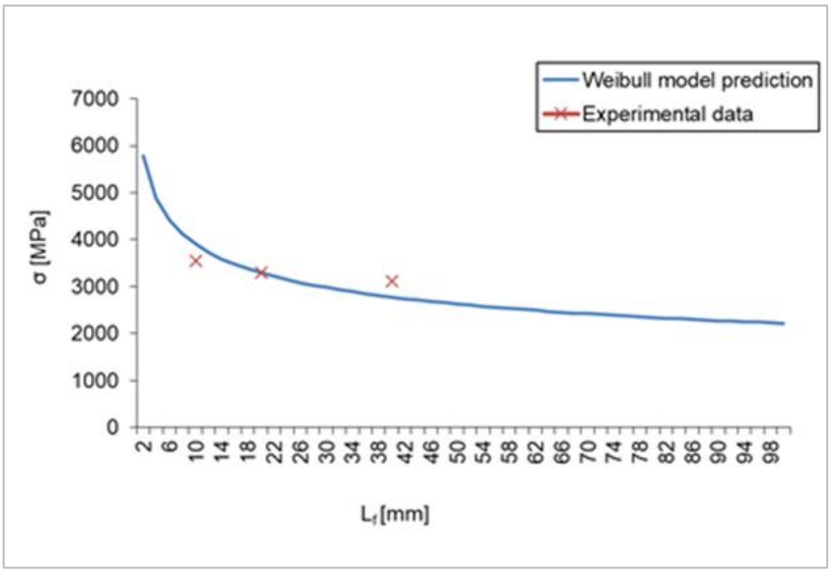


Figure 39: Tensile strength of the sample used as a function of gauge length (reliability = 50%)

Table 18: Properties of rCF, polyester (PL) and polyamide (PA) used for manufacturing ring-spun hybrid yarns

Characteristics	**rCF**	**PL**	**PA**
Fibre diameter (µm)	7.27 ± 0.72	20.1 ± 1.7	18.6 ± 1.5
Single fineness (dtex)	0.46 ± 0.1	3.2 ± 0.1	3.4 ± 0.1
Tensile strength (MPa)	3080 ± 710	423 ± 12	480 ± 23
Young's modulus (GPa)	186 ± 30	3.2 ± 0.18	1.7 ± 0.12
Elongation at break (%)	1.39 ± 0.3	32 ± 5.6	56 ± 7.2
Density (g/cm^3)	1.80	1.38	1.14

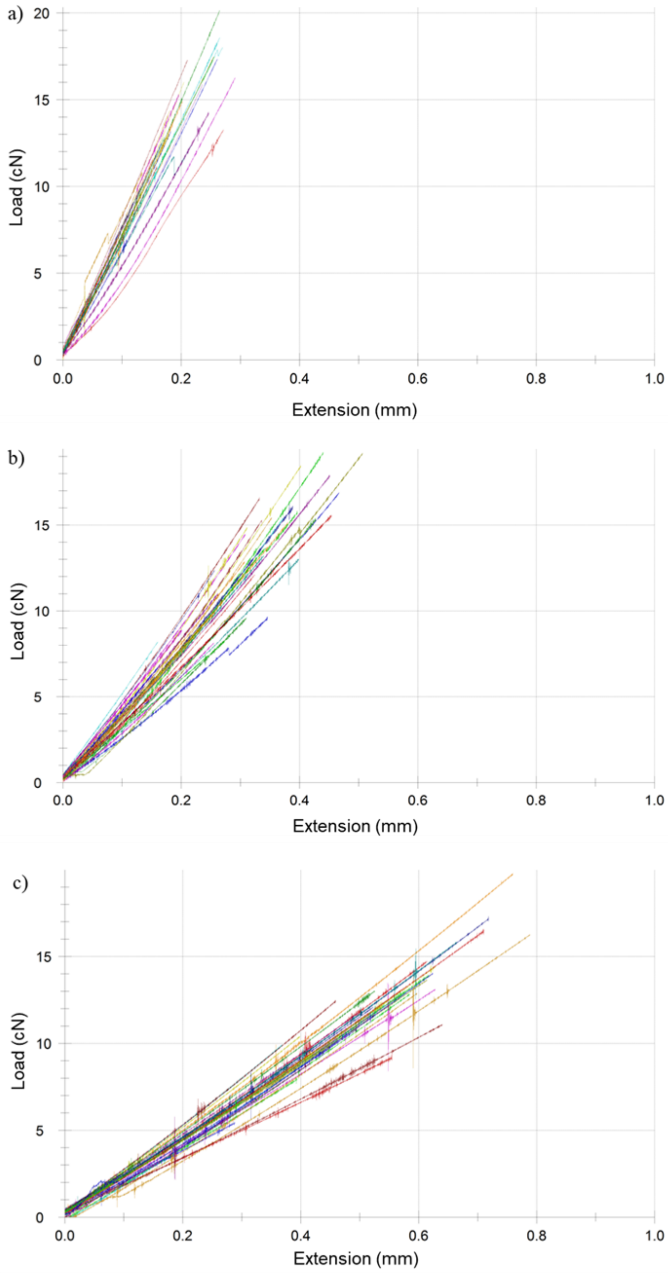


Figure 41: Load-extension curves for 10 (a), 20 (b), and 40 (c) mm gauge length

The values of the tensile strength for the different fibres are reported in Figure 432. Recycled CF is characterised by a clear decrease compared to virgin CF. Specifically, this reduction rises with increasing gauge length (i.e. 28% for 10 mm and 37% for 40 mm). This result is in line with the Weibull statistics analysis represented with Eq. (3).

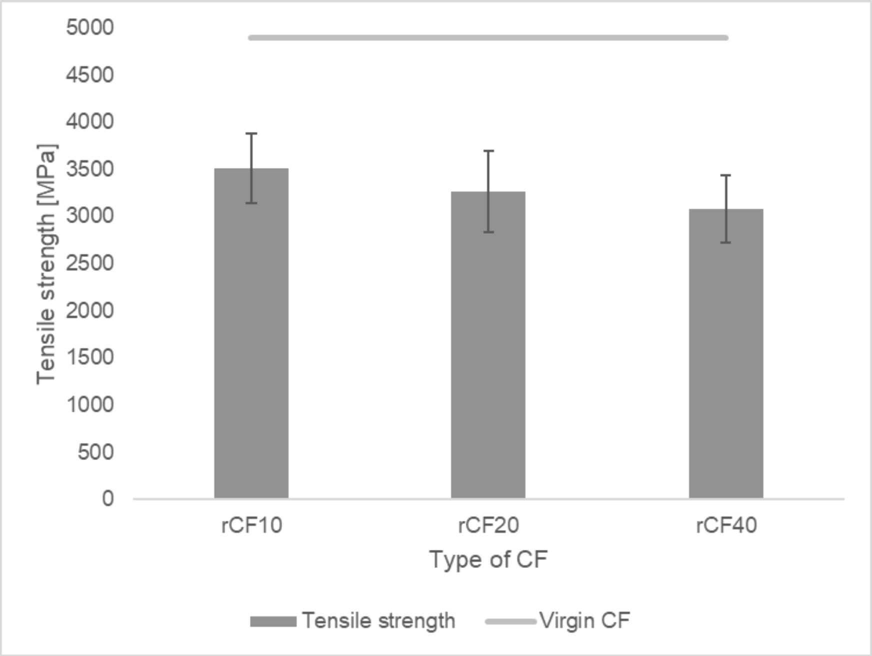


Figure 43: Tensile strength of single filament of recycled CF (standard deviations are shown as error bars)

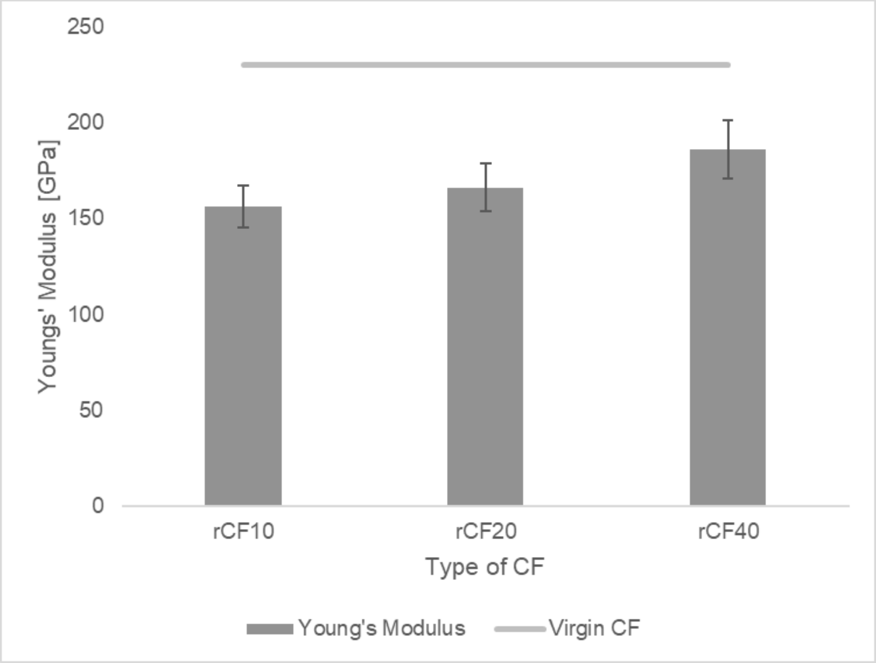


Figure 42: Young’s Modulus of single filament of recycled CF (standard deviations are shown as error bars)

The values of Youngs' Modulus obtained from the mechanical characterisation of single filaments are represented in Figure 423. Recycled CF shows a reduction in modulus equal to 32%, 28% and 19% for gage length of 10, 20 and 40 mm, respectively, compared to virgin CF. Nevertheless, such a decrease may be traced back to the stresses experienced in previous processing when the fibres were in their virgin state.

Visual and quantitative controls

Visual control on the carded webs and carded slivers and quantitative control on the rovings were critical in determining the range of blends of recycled CFs and polyester or polyamide for the fabrication of semi-finished products with an acceptable qualitative level.

The technical feasibility was performed considering three different blending ratios. In particular, 50-50% by weight was chosen as the initial mixture. This choice depended on the fact that the ultimate goal is to make CFRPs that are suitable for structural applications. Indeed, the mechanical properties of the final composite should increase as the CF content increases (Hengstermann et al., 2016b); therefore, evaluating fibre blends with lower CF content would be counterproductive. However, it is also common knowledge that an excessive amount of CF would decrease the mechanical properties of the hybrid yarns and, consequently, the quality of the reinforcement. For these reasons, a right balance should be found. Then, the recycled CFs was increased by 10% until it was no longer possible to produce the roving. Table 19 reports all blends assessed and the results of each checkpoint.

Looking at the quantitative values, it is possible to draw some considerations. First, it may be stated that the proper range of blends that can be processed by the innovative spinning process with the aim of producing hybrid yarns suitable for the production of good-quality CFRPs includes 50%, 60% or 70% recycled CF from manufacturing scraps. The upper limit was defined considering the fact that the mixing ratio of 80% recycled CF and 20% thermoplastic fibre passed the first visual inspection, but not the second. Thus, the higher percentages of CF were not tested.

Second, through the visual assessment and the frequent observations during the carding phase, it can be claimed that the orientation of the CF in the web as well as in the carded sliver is perhaps slightly better for the semi-finished products composed of 50% recycled CF, but still similar for all the percentages considered. As an example, Figure 444 shows the orientation of the CF for the card webs composed of 50% and 70% recycled CF. Such an outcome could be attributed to the fact that the clothing of the carding machine is capable of better processing fibre mixtures in which the percentage of CF is lower. However, as no substantial differences can be perceived, it is not possible to foresee which CFRPs will be the one with the mechanical properties at this stage.

Table 19: Blending ratios and findings of the main control points (PL: polyester and PA: polyamide)

Sample	Visual control I	Visual control II	Quantitative control
rCF-PL3$_{50}$	Passed	Passed	Roving: 43.2%
rCF-PL5$_{50}$	Passed	Passed	Roving: 45.2%
rCF-PA3$_{50}$	Passed	Passed	Roving: 48.8%
rCF-PA5$_{50}$	Passed	Passed	Roving: 48.9%
rCF-PL3$_{60}$	Passed	Passed	Roving: 53.2%
rCF-PL5$_{60}$	Passed	Passed	Roving: 55.7%
rCF-PA3$_{60}$	Passed	Passed	Roving: 57.7%
rCF-PA5$_{60}$	Passed	Passed	Roving: 58.3%
rCF-PL3$_{70}$	Passed	Passed	Roving: 60.8%
rCF-PL5$_{70}$	Passed	Passed	Roving: 65.1%
rCF-PA3$_{70}$	Passed	Passed	Roving: 61.3%
rCF-PA5$_{70}$	Passed	Passed	Roving: 64.3%
rCF-PL3$_{80}$	Passed	Failed	n.a.
rCF-PL5$_{80}$	Passed	Failed	n.a.
rCF-PA3$_{80}$	Passed	Failed	n.a.
rCF-PA5$_{80}$	Passed	Failed	n.a.

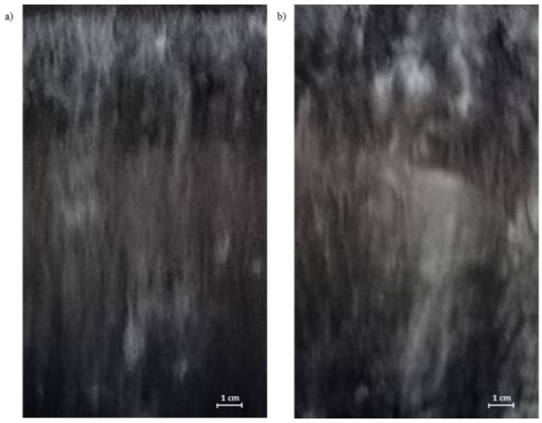


Figure 44: Fibre orientation in the web a) 50-50% rCF and b) 70-30% rCF

Finally, the amount of recycled CF present in the rovings does not correspond to the quantity theoretically entered. In detail, quantitative control shows that on average the process impacts slightly more on blends with higher CF percentages. Likely, in this case, recycled CFs are subjected to more breakages. As a proof of that, Hengstermann et al. (2017) claimed that the carding phase is responsible for a weight loss of 7.9% in the case of 50% recycled CF for hybrid yarns using 60 mm fibre lengths. Furthermore, as the recycled CF percentage increases, the amount of CF remaining in the rovings increases. This result is obviously not surprising, but should not be taken for granted. Indeed, nothing is known about the behaviour between mixed blends and machinery. Specifically, it is not known whether particular chemical-physical or mechanical mechanisms are activated. This point opens up the possibility of further investigating whether the steps in the innovative spinning process influence each blend and, if so, with what intensity. Overall, as the CF reduction within all the rovings produced is acceptable (i.e. less than 10%), all of them were used for the production of hybrid yarns that were then characterised mechanically and thermally and used for the production of thermosetting CFRPs.

Subsequently, the second quantitative control, which corresponds to the characterisation of the physical, thermal and mechanical properties of the hybrid yarns, was carried out. The values of the count for the various hybrid yarns are displayed in Table 20. Specifically, three measurements on a 5 m sample length were made. All in all, findings have relatively high values for variability. This may be attributed to the relatively low technology readiness level (TRL) of the developed innovative spinning process. Thanks to this Ph.D. project, the researcher validated the developed technology at laboratory level; therefore, the TRL was increased from a value of 2 to 3. Moreover, the researcher decided to set at 300 twist per metre the yarn twist of the final spinning machine. Considering the results, it would seem that hybrid yarns made up of 70% recycled CFs have less variability than yarns made up of 50% or 60% recycled CFs. So, it may be said that the whole innovative method is able to better handle blends with greater percentages of recycled CFs. Although the functionality of the innovative process has been tested and established, it definitely merits advancements in the near future to guarantee increased robustness and stability of the final hybrid yarns. Figure 45 reports a ring-spun hybrid yarn example.

Table 20: Value of ring-spun hybrid yarns' count (σ: standard deviation)

Sample	Mean value [dtex]	σ [dtex]
rCF-PL3$_{50}$	580	53
rCF-PL5$_{50}$	447	64
rCF-PA3$_{50}$	640	122
rCF-PA5$_{50}$	680	40
rCF-PL3$_{60}$	630	30
rCF-PL5$_{60}$	424	28
rCF-PA3$_{60}$	681	22
rCF-PA5$_{60}$	698	24
rCF-PL3$_{70}$	501	28
rCF-PL5$_{70}$	500	24
rCF-PA3$_{70}$	507	26
rCF-PA5$_{70}$	695	22

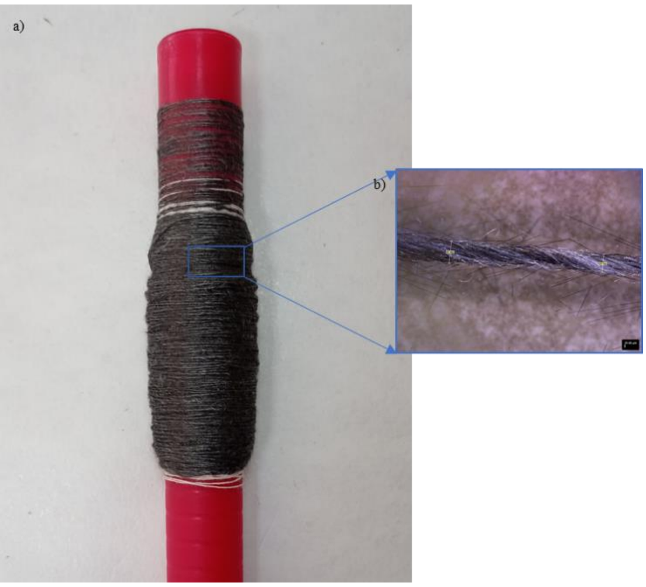


Figure 45: Hybrid yarns manufactured using 50% recycled CF and 50% PA3: a) hybrid yarn bobbin; b) optical microscope image

Figure 466 depicts the results of the TGA. Overall, all the hybrid yarns produced show a loss of recycled CFs probably owing to one or more of the different steps composing the innovative spinning process. This is especially true for hybrid yarns with a 50% recycled CF content, which exhibit the greatest percentage variability. Such a result proves that the whole innovative spinning process has a higher impact on fibre blends with low percentages of recycled CF.

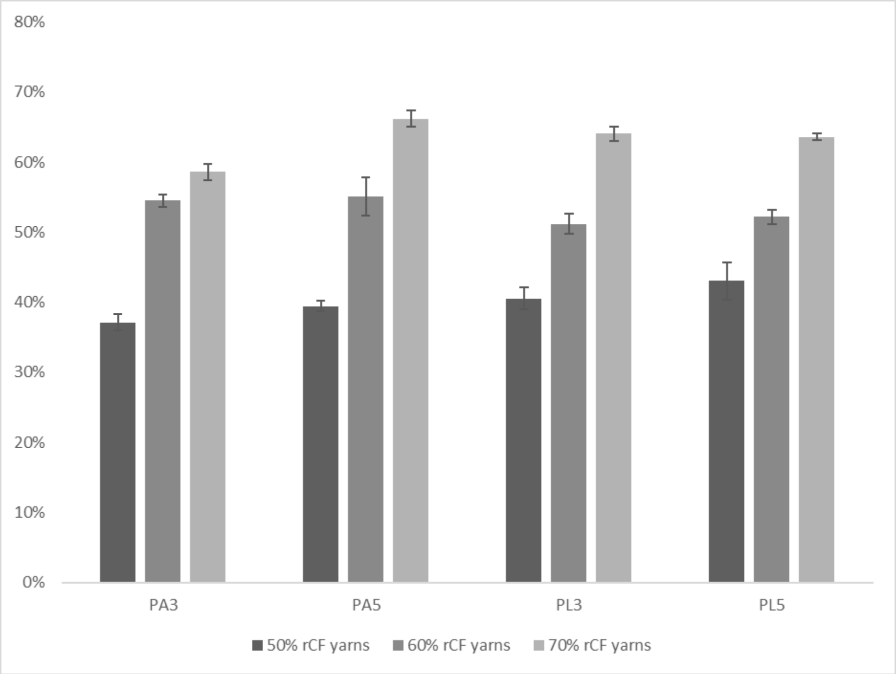


Figure 46: Share of actual amount of recycled CF for all the hybrid yarns produced measured by TGA (standard deviations are shown as error bars)

Table 21 presents the percentage reductions for each hybrid yarn. Looking at the values, it may be stated that, on average, hybrid yarns made from higher quantities of recycled CF are subject to lower reductions. Furthermore, by comparing the percentage reduction on rovings with the percentage reduction on hybrid yarns it is possible to claim that, in general, the ring spinning stage has less impact on the final amount of recycled CF than the carding and drafting stages.

Table 21: Recycled CFs weight percentage measured by TGA at 750°C

Sample	Δ%
rCF-PL3_{50}	9.5
rCF-PL5_{50}	6.9
rCF-PA3_{50}	12.9
rCF-PA5_{50}	10.5
rCF-PL3_{60}	8.9
rCF-PL5_{60}	7.8
rCF-PA3_{60}	5.5
rCF-PA5_{60}	4.9
rCF-PL3_{70}	5.9
rCF-PL5_{70}	6.4
rCF-PA3_{70}	11.4
rCF-PA5_{70}	3.8

Besides, Figure 46 shows that going from 3 to 5 draw frame doubling results in an increase in the percentage of residual recycled CF in all the hybrid yarns regardless of the type of thermoplastic fibre considered. Nevertheless, the most significant increase (i.e. 13%) occurs for hybrid yarn composed of 70% recycled CF and polyamide.

A three-way ANOVA was used to statistically confirm the statements just made. The residuals demonstrated, in all cases, to be normally distributed and randomly scattered with an average value close to zero (Figure 47). Therefore, no evidence exists that the error terms are correlated with one another. The calculated p-values are summarised in Table 22. Overall, it may be claimed that the individual parameters (i.e. recycled CF and number of draw frame doubling) have an impact, in each case in a different way, on the amount of recycled CF that remains after processing. In fact, the low p-values witness that they are statistically significant. Contrariwise, thermoplastic fibre does not seem to have an impact. The same may be claimed for all the interactions apart from the 2-way interaction between recycled CF and the thermoplastic fibre. This could be attributed to potential reactions which may occur when the CF gets in touch with the thermoplastic fibre.

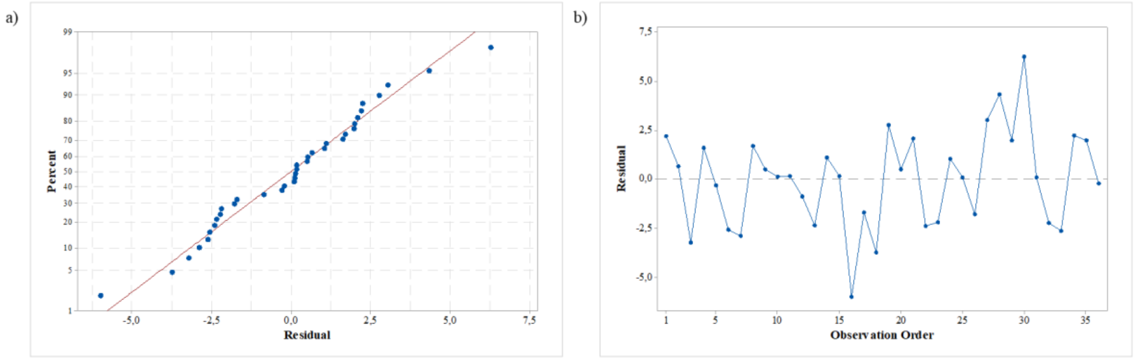


Figure 47: a) Residual normal distribution and b) Randomly scattered residual

Table 22: Three-way ANOVA p-values (p-value: ***<0.01 and **<0.05)

	Factor	**p-value**
Individual parameters	rCF	0.000***
	Thf	0.550
	Ndf	0.033**
2-way interactions	rCF* Thf	0.037**
	rCF* Ndf	0.529
	Thf * Ndf	0.231
3-way interactions	rCF*Thf*Ndf	0.161

Since one of the objectives of the innovative spinning process should be to obtain hybrid yarns with the highest possible content of recycled CF, regardless of the type of thermoplastic fibre, 70% recycled CFs with 5 draw frame doublings appears to be the ideal combination for ring-spun hybrid yarn. This is confirmed also by the main effects plot reported in Figure 48.

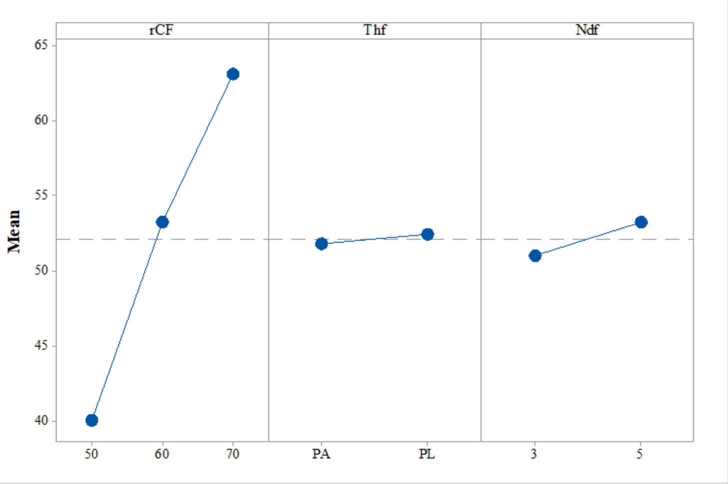


Figure 48: Main effect plot for the output

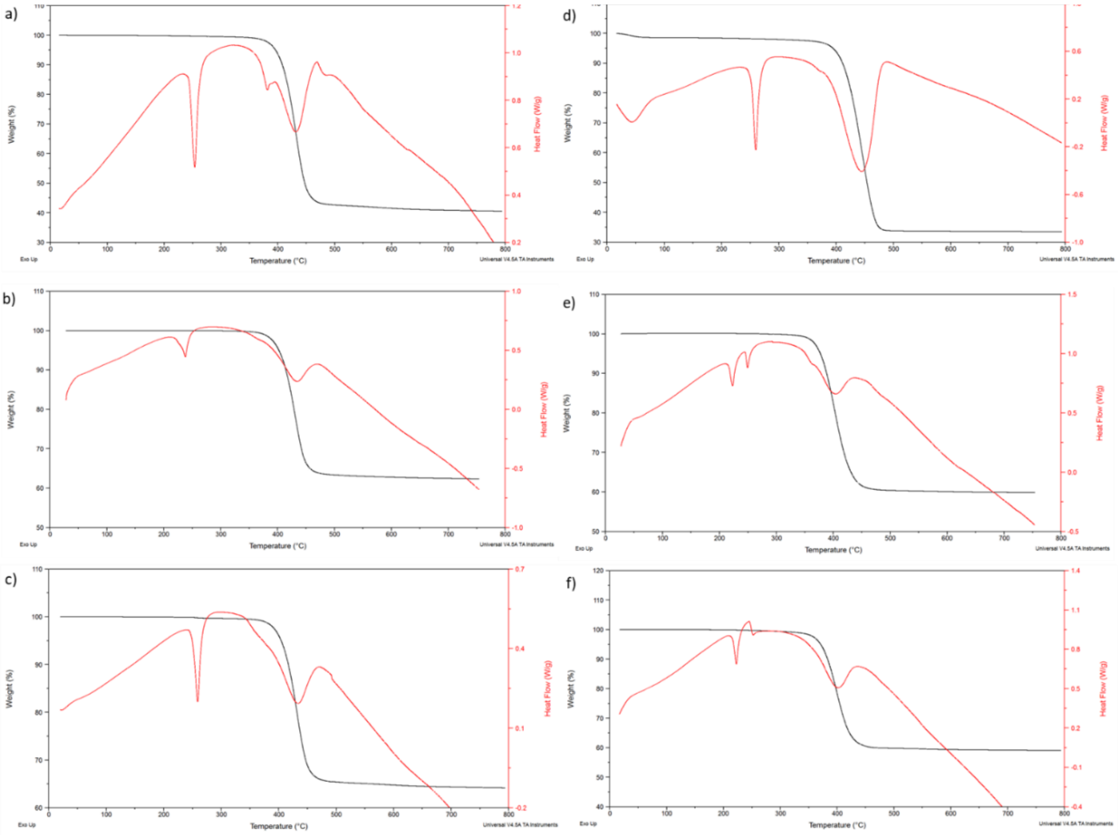


Figure 49: TGA (black) and DSC (red) curves for hybrid yarn a) rCF-PL350, b) rCF-PL360, c) rCF-PL370, d) rCF-PA350, e) rCF-PA360, and f) rCF-PA370

Figure 49 shows, as an example, the thermal behaviour of some of the different hybrid yarns produced. Generally, DSC curve, represented in this case by the red line, may be characterised by both upward and downward peaks. The former represents an exothermal reaction (i.e. a chemical reaction involving a transfer of heat from the system to the environment); while the latter an endothermal reaction (i.e. chemical reaction involving a transfer of heat from the environment to the system). In this investigation, the DSC curves highlight downward trend only. The melting phase, represented by the first peak in the related graph, occurs at approximately 255°C for polyester and at around 260°C for polyamide for all the hybrid yarns. On the other hand, the thermal decomposition of the thermoplastic fibres occurs between 400 and 500°C and is represented by the second deeper peak. This last range of temperatures guarantees that the TGA curves remain stable for temperature values above 500°C. Additionally, since the degradation peak temperature is lower than 450°C for all samples, it may be claimed that the values represented in Figure 46 unquestionably reflect the actual quantity of recycled CFs present in the hybrid yarns.

The outcomes of the tensile tests are listed in Table 23. The values of tenacity depending on recycled CF content and the number of draw frame doubling are shown in Figure 50 a) and b).

Table 23: Tensile properties of the produced ring-spun hybrid yarns

Sample	**Tenacity [cN/tex]**		**Strain at break [%]**		**Young's Modulus [MPa]**	
	Mean	σ	Mean	σ	Mean	σ
rCF-PL3$_{50}$	10.3	3.1	6.5	0.9	917.2	197.4
rCF-PL5$_{50}$	15.1	4.8	6.6	0.7	771.1	149.5
rCF-PA3$_{50}$	8.4	2.7	13.4	2.0	253.5	53.3
rCF-PA5$_{50}$	9.1	3.0	13.1	2.2	278.9	72.7
rCF-PL3$_{60}$	14.4	2.5	3.3	1.0	343.8	83.5
rCF-PL5$_{60}$	24.4	1.4	3.4	0.4	288.1	26.9
rCF-PA3$_{60}$	9.1	2.4	2.7	0.8	272.5	77.0
rCF-PA5$_{60}$	14.8	1.1	4.5	0.7	134.2	10.6
rCF-PL3$_{70}$	13.2	1.2	3.2	0.8	2353.9	399.5
rCF-PL5$_{70}$	10.5	2.0	2.7	1.3	3050.6	609.0
rCF-PA3$_{70}$	6.5	1.4	4.0	0.9	952.5	395.0
rCF-PA5$_{70}$	13.5	2.4	2.0	0.2	2348.1	542.5

It may be stated that the tenacity of ring-spun hybrid yarns increases passing from a number of draw frame doubling equal to 3 to 5 for both types of thermoplastic fibres. For instance, considering 50% recycled CF content, raising the number of draw frame doubling the tenacity increases by 8.3% and 46.6% for polyamide and polyester, respectively. Instead, when considering 60% recycled CF content, the increase is significant for both thermoplastic fibres, but higher for polyester (69.4% compared to 62.6% for polyamide). However, the largest increase (i.e. 107.7%) is observed for hybrid yarns consisting of 70% recycled CF and polyamide. The only hybrid yarn that shows a decrease in tenacity (around 20.5%) from 3 to 5 of draw frame doubling is that composed of polyester blended with 70% recycled CF.

Contrariwise, it appears that the recycled CF content does not systematically increase the tenacity of ring-spun hybrid yarns. This outcome is not unexpected, as the likelihood of fibres slipping on each other and reducing strength may rise with the increase in the amount of recycled CF in the yarn. Furthermore, the thermoplastic fibre considered may also play a crucial role. Indeed, polyester-based ring-spun hybrid yarns generally outperformed polyamide-based ones in terms of tensile strength. This result could be explained by possible interactions between the thermoplastic fibre under study and the recycled CF.

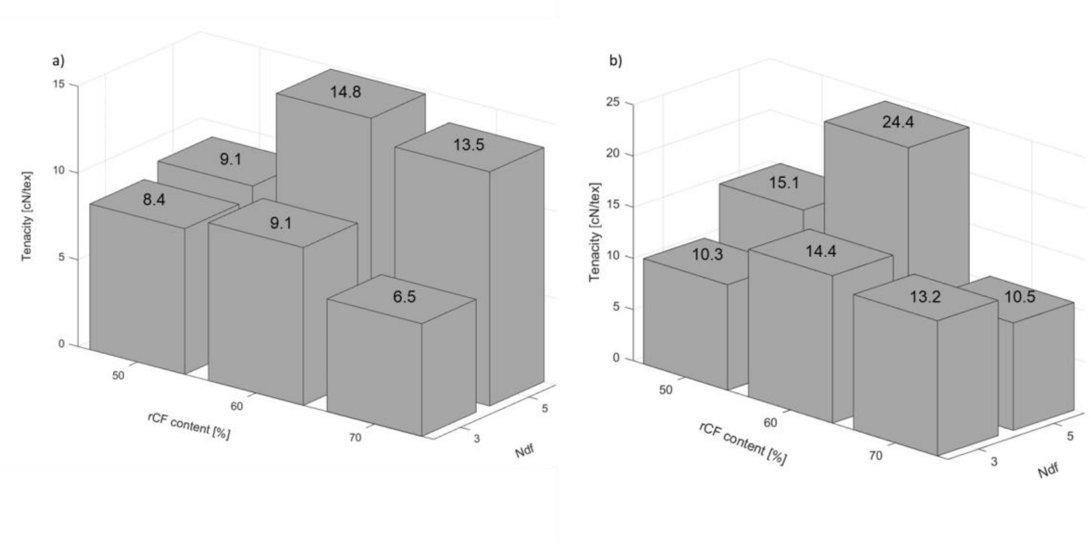


Figure 50: Tenacity of ring-spun hybrid yarns considering recycled CF content and number of draw frame doubling for a) polyamide and b) polyester fibres

Lastly, it is possible to state that as the recycled CF content increases, yarns become less ductile. For instance, ring-spun hybrid yarns composed of 70% recycled CF are characterized by higher tenacity but lower strain at break than those composed of 50% recycled CF. This finding is supported by the fact that 70% recycled CF yarns exhibit noticeably higher Young's modulus values. Therefore, due to the likelihood of the recycled CF slipping over each other and the reduced amount of thermoplastic fibres functioning as carriers, breakages occur fairly easily in the case of 70% recycled CF.

Additionally, Figure 51 displays the tensile strength of the ring-spun hybrid yarns in relation to the actual amount of recycled CF. Looking at the values, it may be stated that the higher the actual amount of remaining recycled CF the higher the tensile strength and vice versa. All in all, regardless of the type of thermoplastic fibre at hand, the actual amount of recycled CF is higher for hybrid yarns composed of a number of draw frame doubling equal to 5, thus resulting in higher values of tensile strength. Such a result could be attributed to the fact that in this case the drawing phase is gentler and increases fibre parallelisation to a greater extent.

Finally, an assumption regarding the mode of breakage of hybrid yarns can be extracted. More in detail, by applying a constant load, it is possible to state that the yarn starts to unravel until breakage occurs (Figure 52). Therefore, it appears that the thermoplastic fibre breaks first, followed by the CF. It can hence be stated that the yarn continues to resist due to the presence of the latter. Obviously, more specific analyses should be performed to provide precise consideration on this topic.

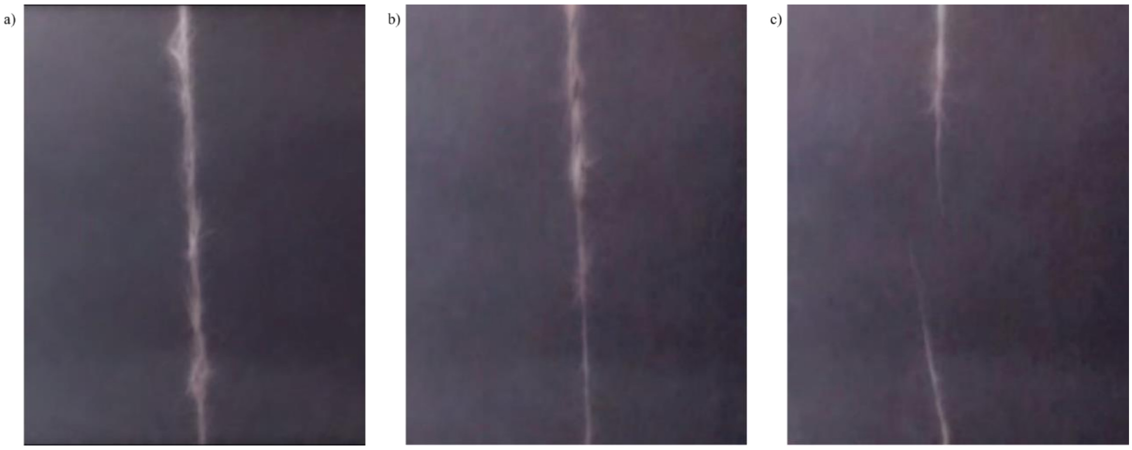


Figure 51: Breaking mode of a 70% RCF and 30% polyester yarn with 3 as the number of draw frame doublings

3.6 Conclusions

This Chapter proves the technical feasibility of the innovative spinning process for the production of ring-spun hybrid yarns using recycled CF from manufacturing scraps blended with virgin polyester or polyamide fibres. Bearing the final goal in mind, it was possible to identify that the quantity of recycled CF allowing a good balance between tensile strength and effective amount of recycled CF ranges between 50 and 70% by weight.

Overall, outcomes highlight that fibre orientation is crucial especially for the card web, whereas blending ratio for the whole process. In detail, webs produced with an amount of recycled CF equal to 50% have better fibre orientation as card clothing on the carding machine works better with little quantities of recycled CF.

Moreover, regardless of the type of thermoplastic fibre, the number of draw frame doubling seems to affect the tenacity of ring-spun hybrid yarns. Indeed, the tensile tests show that tenacity rises, passing from 3 to 5. Contrariwise, the increase in recycled CF content does not strongly impact the tenacity of ring-spun hybrid yarns. However, the actual amount of remaining recycled CF appears to be correlated with the tensile strength value. Specifically, ring-spun hybrid yarns composed of 70% of recycled CFs suffer from a lower decrease in the quantity of recycled CF and have inferior percentages for both polyester and polyamide fibres. Accordingly, they are generally characterized by slightly higher tensile strength values. All in all, ring-spun hybrid yarns composed of 70% recycled CF, possess the best mechanical and thermal properties, regardless of the type of thermoplastic fibre.

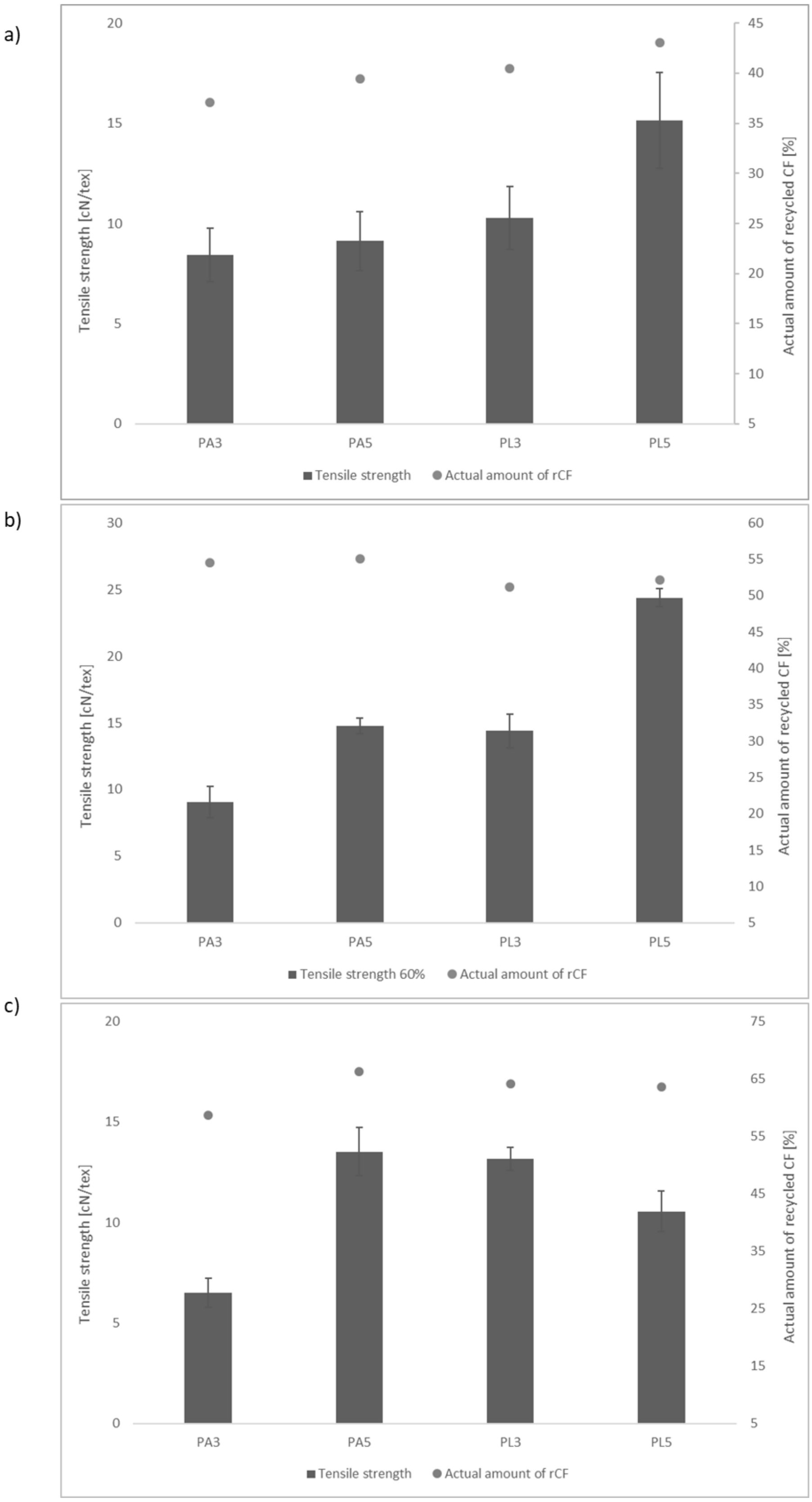


Figure 52: Tensile strength for the hybrid yarns composed of a) 50%, b) 60%, and c) 70% rCFs with respect to the real amount of remaining rCFs. The left y-axis represents the tensile strength [cN/tex], while the right y-axis represents the actual remaining amount of rCFs [%] (standard deviations are shown as error bars)

CHAPTER 4

Production and characterisation of carbon fibre-reinforced plastic composite materials

4.1 Introduction

Virgin CFRP composite materials exhibit several advantages. First of all, they are characterised by outstanding mechanical and physical properties, such as high specific strength, high specific stiffness and low density, but also by durability in aggressive conditions and at high temperatures, abrasion resistance and corrosion resistance (Clark et al., 2020; Hadigheh et al., 2020; Zhang et al., 2020). Over the recent years, the demand for these materials has drastically increased also owing to CO_2 emission reduction goals and the need to lighten vehicle constructions (Zhang et al., 2020). In such a context, worldwide CFRP waste is expected to reach 20 kt/year by 2025 (Rademacker, 2018). Accordingly, several challenges emerge on waste management especially from an environmental perspective. The recovery of CF through recycling technologies or reusing and subsequent remanufacturing practices is environmentally attractive, particularly in light of circular economy. Indeed, it would avoid landfilling and incineration, which, over the last years, have been the most common disposal approaches for end-of-life CFRPs (Oliveux et al., 2015), as well as the high emission of greenhouse gases due to the production of new virgin CF (van de Werken et al., 2019).

To overcome potential significant environmental issues and align with circular economy principles, researchers are trying to develop new remanufacturing methods for recycled CFRPs (van de Werken et al., 2019). So far, research has focused on the study of methods that are typical of discontinuous fibre composites for the production of recycled CFRPs. Outcomes are represented by randomly oriented or uniformly aligned composites which usually do not exhibit remarkable mechanical properties. Some scientific studies have drawn attention to the importance of fibre alignment (e.g. Turner et al., 2013; Yu et al., 2014) for improving the mechanical properties of the recycled CFRP composites and an overview of techniques suitable for fibre alignment is proposed by Such et al. (2014).

Hydrodynamic techniques allow the highest degree of alignment (Longana et al., 2021). Among them, the most common known are rotating drum developed at the University of Nottingham (Wong et al., 2010), HiPerDiF from the University of Bristol and Lineat Composites (Longana et al., 2016) and TuFF process by the University of Delaware (Tierney et al., 2019). The latest recognised method for improving the alignment of CF, which has recently attracted the interest of researchers, is the spinning process.

In such a context, due to their compactness and high fibre orientation, yarns currently represent a great opportunity for producing value-added products (Hengstermann et al., 2016b; Pakdel et al., 2020). Some attempts have already been made to manufacture recycled CFRPs. For instance, Hengstermann et al. (2016) and Hengstermann et al. (2017) fabricated unidirectional thermoplastic CFRPs by using a laboratory press machine P 300 V (Dr. Collin GmbH, Germany). Consolidation was performed by an under vacuum computer-controlled cycle following a heating step ranging between 30-280°C and varying the pressure. Results of Hengstermann et al. (2016) showed that among all yarns the one made from 80 mm CFs and PA6 fibres provides the highest tensile strength of 800 MPa to the unidirectional composite. As regards Hengstermann et al. (2017), instead, the highest tensile strength (i.e. 838 MPa) was achieved by the hybrid yarn composed of 60 mm CFs and PA6 in the case of 87 T/m containing 50% volume of CFs.

To the best of researcher’s knowledge, there are no studies on the realisation of unidirectional thermosetting composite materials. Therefore, in this Chapter a first attempt for the production and characterisation of unidirectional thermosetting composite materials reinforced with recycled CFs is presented. The ultimate aim is to understand which of the different samples made at laboratory level has the best mechanical characteristics in terms of tensile strength and elastic modulus. Hybrid yarns produced through the innovative spinning process, as described in Chapter 3 of this manuscript, were used for this purpose. The findings of this explorative analysis could pave the way for future research focused on a more in-depth analysis of the mechanical properties of the CFRP that appears to be most promising in the current study.

4.2 Materials and methods

4.2.1 Materials

Ring-spun hybrid yarns produced through the innovative spinning process were used as reinforcement. Their physical and mechanical properties have been already determined. For further information, please refer to Section 3.5 of Chapter 3 of this manuscript.
An epoxy resin was used as the matrix. Specifically, a cold curing epoxy system was adopted. Such a system is composed of Araldite® LY 5052, a low viscosity epoxy resin, and Aradur® 5052, a mixture of polyamines to harden the resin. Both these components were supplied by HUNTSMAN. The epoxy system has numerous uses in tooling, aviation maintenance, industrial and aerospace composites. According to the datasheet of the manufacturer, at 25°C, Araldite® LY 5052 has a density of 1.17 g/cm^3, while Aradur® 5052 of 0.94 g/cm^3.

4.2.2 Methods

4.2.2.1 Specimen specifications

In order to investigate the tensile properties of unidirectional thermosetting composite materials reinforced with recycled CFs, particular specimens were produced. In detail, a dog-bone shaped specimen was selected. The choice fell on this type of shape for two main reasons. First, several standards exist for the preparation and subsequent characterisation of composites either thermoplastic or thermosetting reinforced with multiple layers of unidirectional reinforcements arranged at different angles (i.e. ISO 527-4:1997) or a single unidirectional fabric (i.e. ISO 527-5:1997). However, they all involve the use of virgin CFs. Furthermore, the topic at hand is significantly innovative and experimental; therefore, a well-known standard to follow is not available. Second, it has been specifically designed to concentrate the breaking stress in the central part of the specimen. **Figure 53**Figure 53 shows the geometry of the test specimens used for tensile

Figure 53: Geometry of the test specimens used

testing. The length is 80 mm, the width at the centre of the specimen is 5 mm and the thickness is 2 mm.

4.2.2.2 Preparation of the reinforcement

In order to improve the performance of the final CFRPs, hybrid yarns were placed side by side to produce a unidirectional textile substrate. Such a unidirectional textile substrate was realised by manually wrapping five times the hybrid yarn on a wrapping frame developed by the researcher for this project. As it is crucial that the shape does not change during the wrapping phase, the wrapping frame was produced by 3D printing.

The realisation of the wrapping frame required four phases:

1. **Design**: A sketch on paper was essential to define the shape of the wrapping frame in the correct way. Indeed, this is phase allowed the researcher to cope with the different requirements for the realisation of an element that was useful for the final purpose. Obviously, specimen measurements were considered.

2. **CAD modelling**: SOLIDWORKS®, a parametric modelling software, was used for the realisation of the 3D model of the wrapping frame (Figure 54). Once the 3D model was made and the material defined, the print was sent out.

Figure 54: The 3D model of the wrapping frame

3. **Printing**: To print the wrapping frame, an Onyx Pro^{TM} (Figure 55) machine supplied by Markforged[4] was employed. The Onyx Pro^{TM} is an advanced professional 3D printer leveraging Continuous Fibre Reinforcement (CFR) technology to reliably produce robust components.

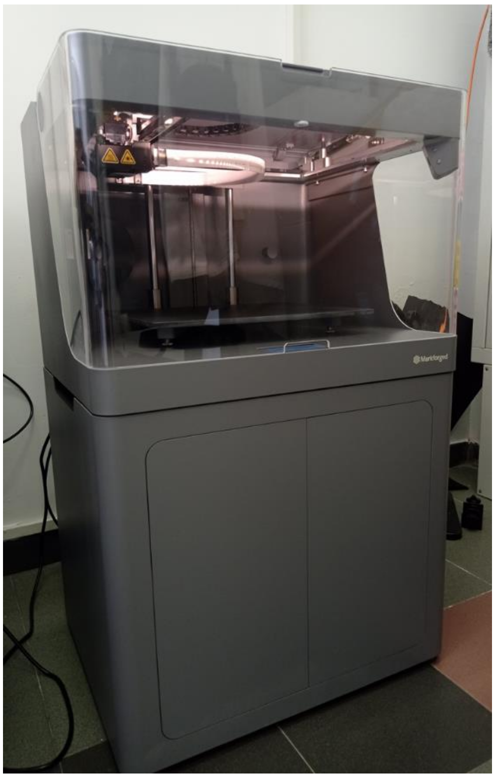


Figure 55: The 3D printing machine adopted for the realisation of the wrapping frame

4. **Cleaning:** To make the part, the 3D printer needs little supports, which are made automatically. However, these supports, once the part is finished, need to be removed in order to proceed with the correct use of the part.

The wrapping frame was realised through a carbon microfibre-filled nylon filament. An example of the wrapping frame produced is shown in Figure 56.

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[4] https://markforged.com

Figure 56: Example of the wrapping frame realised through 3D printing

4.2.2.3 Preparation of the unidirectional thermosetting CFRPs

First of all, the silicone mould was dried at 80°C for 1 hour in order to remove any humidity. Second, the unidirectional textile substrate was placed inside the silicone mould and clamped into the ends of the specimen by means of a stapler. Then, the matrix was prepared. In detail, in accordance with the datasheet, the resin and hardener were mixed in a proportion of 100:38 by weight. After being stirred through a magnetic stirrer, the matrix was subjected to a de-gassing process for 15 minutes to minimise the presence of air. At this point, it was poured into the cavity through a pro-pipette taking care not to allow air bubbles to form and to ensure that the reinforcement remains inside the specimen. Subsequently, the silicone mould was inserted in the oven to activate the matrix curing process. The dwell time in the oven was characterised by a double cycle. Initially, the temperature was set at 80°C for 1 hour. Then, the temperature was raised to 150°C and curing of the specimen continued for a further two hours. Finally, after drying the unidirectional thermosetting CFRP, potential excess resin was removed and the

Figure 57: Example of the specimen produced

composite was filed down by means of a grinding wheel. An example of the specimen of unidirectional thermosetting CFRP manufactured is depicted in Figure 57.

4.2.2.4 Mechanical test

All mechanical tests were performed at room temperature. An MTS ALLIANCE RT50 2 kN dynamometer equipped with self-tensioning wedge grips was used. In detail, five repetitions for each batch considered were carried out, at a constant crosshead rate of 0.2 mm/min and with a distance between the grippers of 58 mm. The elastic modulus, the tensile strength and the elongation were computed using Testworks®4 software considering the measurements of the specimen. However, to measure the exact extension of the material under load, a contact extensometer was adopted, specifically a sensor arm extensometer, whose gage length was set to 10 mm (Figure 58).

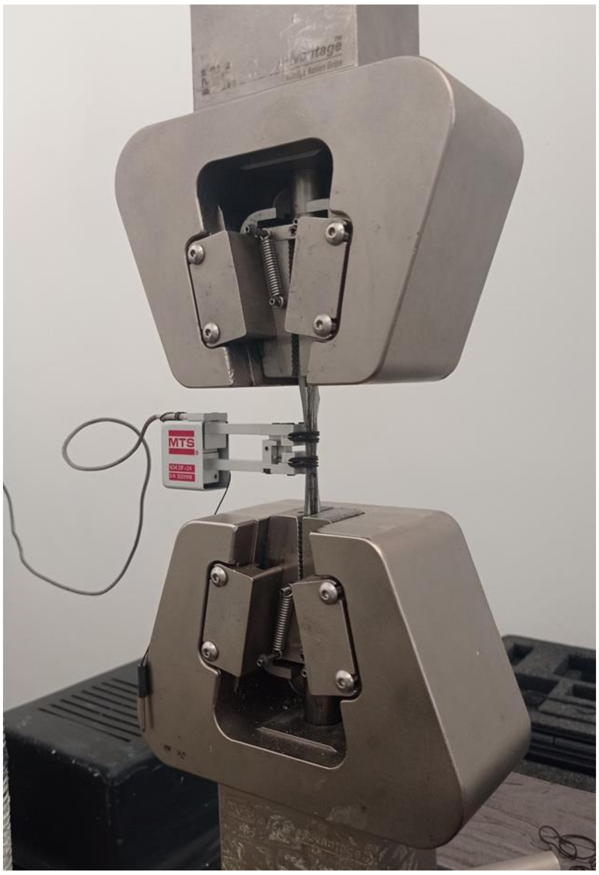

Figure 58: Self-tensioning grips and MTS® contact extensometer used

4.2.2.5 Plan of experiments

The experimental campaign was conducted on the base of a 3 x 2 x 2 full factorial design of experiments (DoE) by varying the amount of recycled CFs (rCF) on three levels, while the type of thermoplastic fibre (ThF) and the number of draw frame doubling (Ndf) on two levels (low and high). Five repetitions were considered for each combination of the parameters. Table 24 shows the 3 x 2 x 2 factorial plan displaying the combination of parameters. The main characteristics of the ring-spun hybrid yarns produced through the innovative spinning process were considered to define the values of the levels of each parameter. They are reported in Table 25.

The analysis of variance (ANOVA) was adopted to verify their potential influence on the mechanical response properties (i.e. Young's Modulus and Tensile strength) of the produced unidirectional thermosetting CFRPs. To this end, a confidence interval equal to 95% was considered.

Table 24: Design of experiments - Parameters combinations

Samples	rCF	ThF	Ndf
1-13-25-37-49	50	PA	3
2-14-26-38-50	60	PA	3
3-15-27-39-51	70	PA	3
4-16-28-40-52	50	PL	3
5-17-29-41-53	60	PL	3
6-18-30-42-54	70	PL	3
7-19-31-43-55	50	PA	5
8-20-32-44-56	60	PA	5
9-21-33-45-57	70	PA	5
10-22-34-46-58	50	PL	5
11-23-35-47-59	60	PL	5
12-24-36-48-60	70	PL	5

Table 25: Parameters and related levels

Parameters	Abbreviation	Low level	Medium level	High level
Amount of recycled CF [%]	rCF	50	60	70
Thermoplastic fibre	ThF	PA	-	PL
Number of draw frame doubling	Ndf	3	-	5

4.3 Results and discussion

4.3.1 Tensile strength

A three-way ANOVA was used to statistically assess the potential influence of the different factors considered on the tensile strength of the final composite specimens.

The residuals demonstrated, in all cases, to be normally distributed and randomly scattered with an average value near to zero (Figure 59).

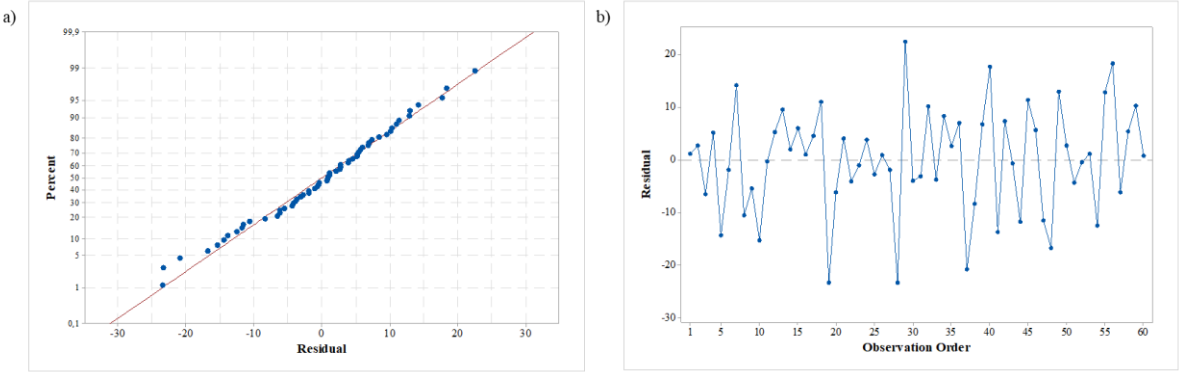

Figure 59: a) Residual normal test and b) Randomly scattered residual

No evidence heteroscedasticity emerges from the sample; thus, the assumptions of the ANOVA are met. The calculated p-values are summarised in Table 26.

Table 26: Three-way ANOVA p-values (p-value: ***<0.01 and **<0.05)

	Factor	**p-value**
Individual parameters	rCF	0.000***
	Thf	0.911
	Ndf	0.019**
2-way interactions	rCF* Thf	0.001***
	rCF* Ndf	0.009***
	Thf * Ndf	0.013**
3-way interactions	rCF*Thf*Ndf	0.245

The percentage of recycled CF and the number of draw frame doubling of the hybrid yarns have an impact, in each case in a different way, on the tensile strength of the thermosetting CFRP composite materials. In fact, the low p-values witness that their influence is statistically significant. Contrariwise, thermoplastic fibre does not have an impact. The same may be claimed for the 3-way interaction. On the other hand, all the 2-way interactions seem affect the tensile strength of the final CFRPs produced (Figure 60).

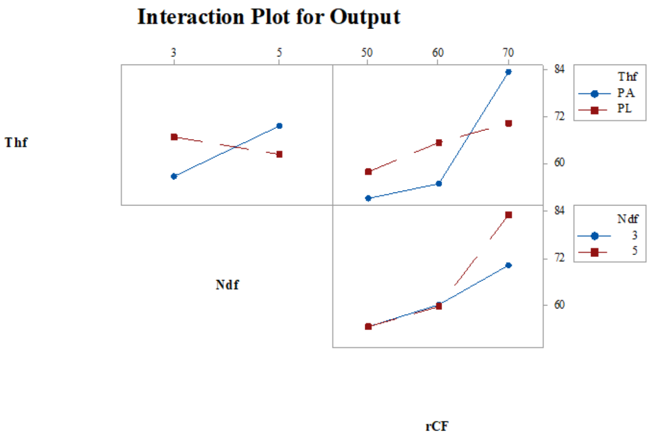

Figure 60: Interaction plot for the outputs

Since the main purpose is to produce thermosetting CFRPs with the best mechanical properties, regardless of the type of thermoplastic fibre, 70% recycled CFs with 5 draw frame doublings appears to be the ideal combination. This is confirmed also by the main effects plot reported in Figure 61.

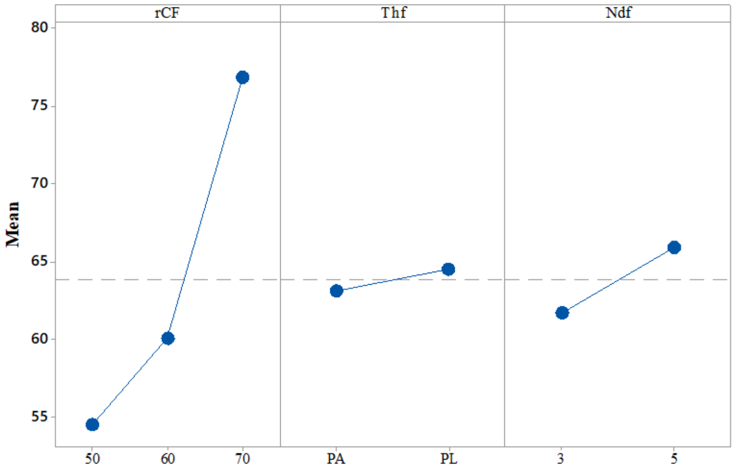

Figure 61: Main effects plot for the output

The tensile strength of all the thermosetting CFRP composites and of the neat epoxy resin, taken as a reference, is represented in Figure 62.

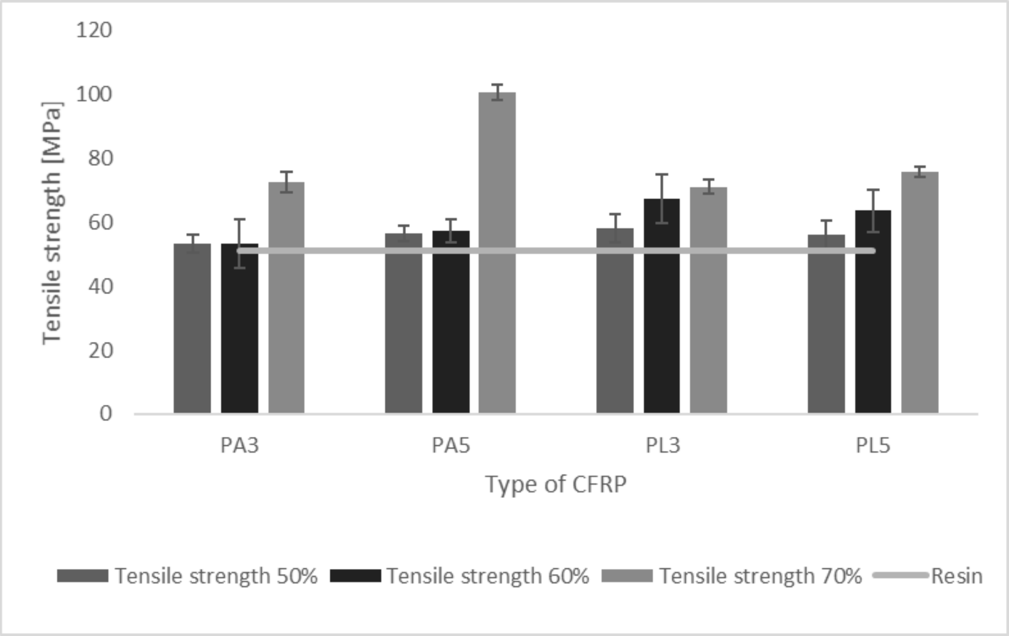

Figure 62: Tensile strength of all thermosetting composites produced compared to neat resin (standard deviations are shown as error bars)

Overall, the tensile strength increases when increasing the percentage of recycled CF present, as well as the number of draw frame doubling, regardless of the type of thermoplastic fibre used.

As expected, all CFRP composites consisting of the hybrid yarns previously produced were clearly stronger than the neat epoxy matrix, which exhibits a tensile strength of approximately 50.9 MPa. All in all, the CFRP composed of rCF-PA3$_{50}$ hybrid yarn shows the lowest tensile strength with 53.3 MPa. Slightly higher values could be achieved in composites reinforced with hybrid yarns consisting of 60% of recycled CF. Whereas the CFRP that exhibits the highest tensile strength (i.e. 100.6 MPa) is composed of rCF-PA5$_{70}$ hybrid yarn.

Lastly, Figure 63 shows the improvement in tensile strength of the thermosetting CFRPs when compared to the neat epoxy matrix. In detail, the highest increase was 97.7% and was achieved by the composite material consisting of hybrid yarns made up of 70% recycled CF and 30% polyamide with a number of draw frame doubling is equal to 5. In contrast, the lowest increase (i.e. 4.72%) was achieved by the composite material consisting of hybrid yarns made up of 60% recycled CF and 40% polyamide with a number of draw frame doubling is equal to 3.

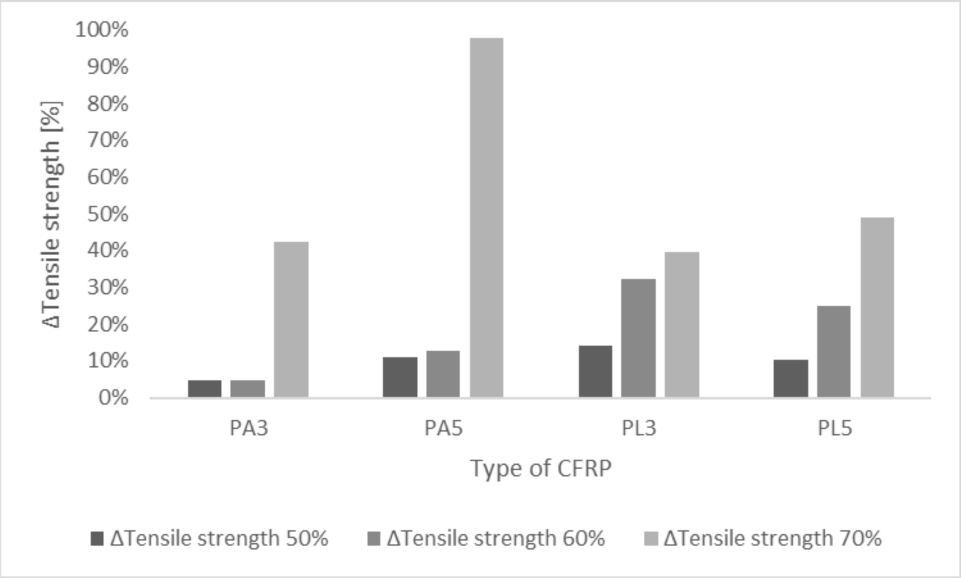

Figure 63: Improvement in tensile strength for each CFRP composed of hybrid yarns with 50%, 60%, and 70% recycled CF compared to tensile strength of neat resin

4.3.2 Young's Modulus

A three-way ANOVA was used to statistically assess the potential influence of the different factors considered on the Young's Modulus of the final composite specimens.

The residuals demonstrated, in all cases, to be normally distributed and randomly scattered with an average value near to zero (Figure 64).

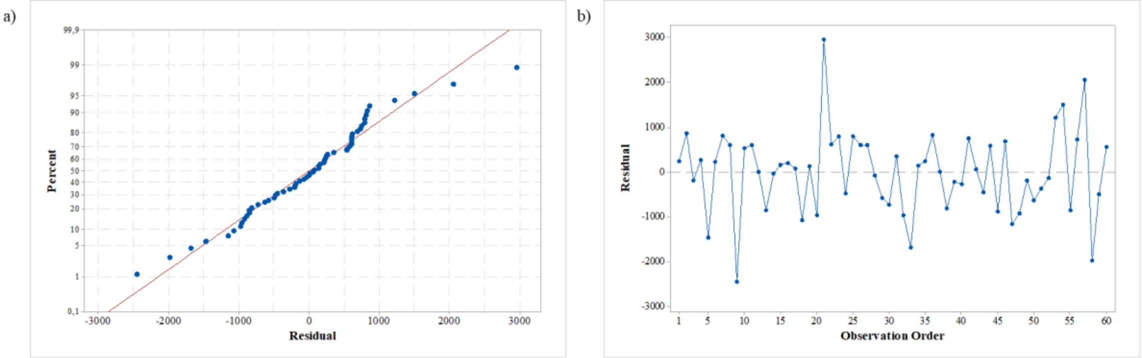

Figure 64: a) Residual normal test and b) Randomly scattered residual

No evidence heteroscedasticity emerges from the sample; thus, the assumptions of the ANOVA are met. The calculated p-values are summarised in Table 27.

Table 27: Three-way ANOVA p-values (p-value: ***<0.01 and **<0.05)

	Factor	p-value
Individual parameters	rCF	0.000***
	Thf	0.602
	Ndf	0.041**
2-way interactions	rCF* Thf	0.975
	rCF* Ndf	0.036**
	Thf * Ndf	0.445
3-way interactions	rCF*Thf*Ndf	0.919

The percentage of recycled CF and the number of draw frame doubling of the hybrid yarns have an impact, in each case in a different way, on the Young's Modulus of the thermosetting CFRP composite materials. In fact, the low p-values witness that that they are statistically significant. Contrariwise, thermoplastic fibre does not have an impact. The same may be claimed for all the interactions apart from the 2-way interaction between recycled CF and the number of draw frame doubling (Figure 65).

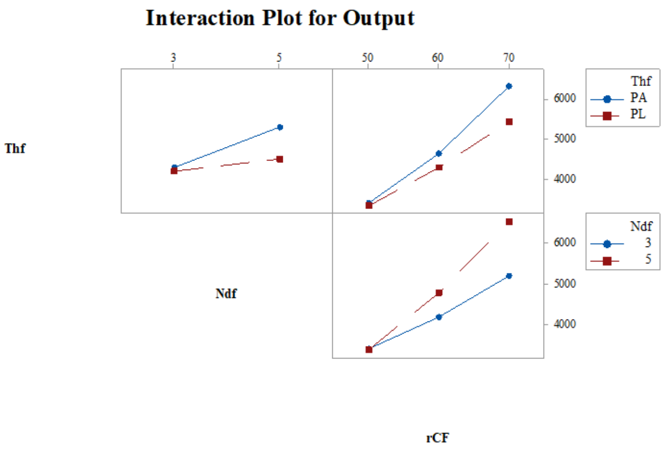

Figure 65: Interaction plot for output

Since the main purpose is to produce thermosetting CFRPs with the best mechanical properties, regardless of the type of thermoplastic fibre, 70% recycled CFs with 5 draw frame doublings appears to be the ideal combination. This is confirmed also by the main effects plot reported in Figure 66.

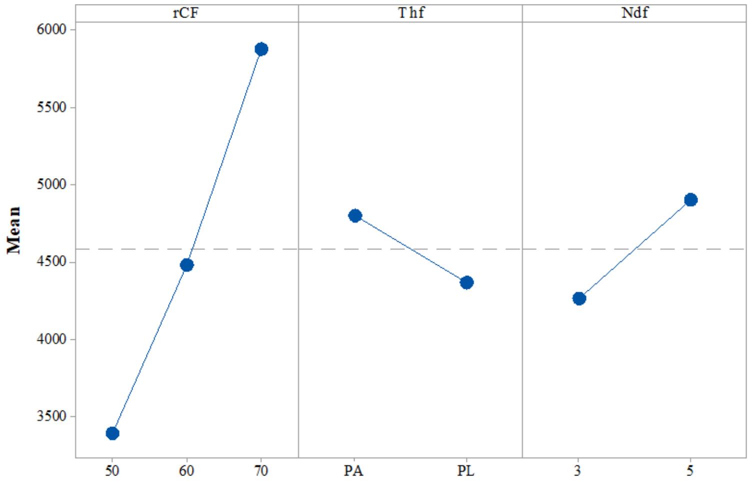

Figure 66: Main effects plot for the output

The Young’s Modulus of all the thermosetting CFRP composites and of the neat epoxy resin, taken as a reference, are represented in Figure 67.

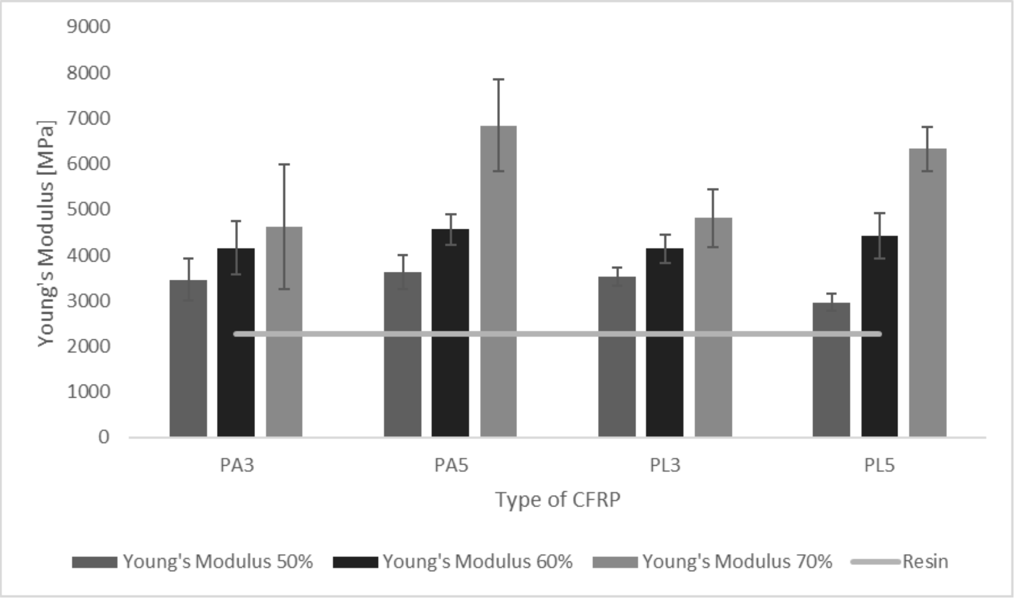

Figure 67: Young's Modulus of all thermosetting composites produced compared to neat resin (standard deviations are shown as error bars)

It is generally recognised that the modulus of a composite rides on the moduli and weight fraction of the composite constituents (F. Wang et al., 2015). In this case, the Young’s Modulus increases when increasing the percentage of recycled CF present, as well as the number of draw frame doubling, regardless of the type of thermoplastic fibre used.

As expected, all CFRP composites consisting of the hybrid yarns previously produced were characterised by a higher level of stiffness than the neat epoxy matrix, which exhibits a Young's Modulus of approximately 2271.2 MPa. All in all, the CFRP composed of rCF-PL5$_{50}$ hybrid yarn shows the lowest Young's Modulus with 3029.1 MPa. Slightly higher values could be achieved in composites reinforced with hybrid yarns consisting of 60% of recycled CF. Whereas the CFRP that exhibits the highest Young's Modulus (i.e. 6846.8 MPa) is composed of rCF-PA5$_{70}$ hybrid yarn.

The theoretical Young's Modulus of all the thermosetting CFRPs produced was calculated according to the rule of mixtures Eq. (6) (Moser, 1992).

$$E_c = E_f . \varphi_f + E_m . \varphi_m \tag{6}$$

where E_f is the Young's Modulus of the hybrid yarn at hand and E_m the Young's Modulus of the matrix, while φ_f, φ_m are the volume content of the hybrid yarn and the matrix volume content of the composite under investigation.

Specifically, the actual surface area of the yarn from the count was considered to compute φ_f, and tabulated values equal to 1.14 g/cm^3 and 1.38 g/cm^3 (ISO 1183) were used for the density of polyamide and polyester, respectively. While the density of CF is 1.17 g/cm^3 from the technical datasheet. Finally, the Young's Modulus of the matrix was set to 3000 MPa from the datasheet and of the single rCF to 186 GPa based on the Single fibre tensile tests previously performed. To calculate the Young's Modulus of each hybrid yarn, the volume fraction of rCF was computed from the volume of the hybrid yarn. Then, the volume fraction of the thermoplastic fibre was assimilated into the matrix. Obviously, this is a first approximation. To obtain more precise results, it would be appropriate to consider the thermoplastic fibre as a self-standing phase.

Findings show how the measured values are higher than the calculated values according to the rule of mixtures (Eq. (6)) by approximately 49% (Table 28). This may be attributed to an irregular surface in the central area of the specimens.

Lastly, Figure 68 shows the improvement in Young's Modulus of the thermosetting CFRPs when compared to the neat epoxy matrix. In detail, the highest increase was

201.5% and was achieved by the composite material consisting of hybrid yarns made up of 70% recycled CF and 30% polyamide with a number of draw frame doubling is equal to 5. In contrast, the lowest increase (i.e. 30.5%) was achieved by the composite material consisting of hybrid yarns made up of 50% recycled CF and 50% polyamide with a number of draw frame doubling is equal to 5.

Table 28: Comparison of measured and calculated Young's Modulus in MPa of the investigated composites

Composite	Young's Modulus calculated with the rule of mixtures	Measured Young's Modulus
rCF-PL3$_{50}$	3279.8	3536.1 ± 392
rCF-PL5$_{50}$	3215.5	3029.1 ± 265
rCF-PA3$_{50}$	3377.3	3463.5 ± 913
rCF-PA5$_{50}$	3400.9	3628.2 ± 767
rCF-PL3$_{60}$	3356.3	4137.4 ± 636
rCF-PL5$_{60}$	3240.2	4419.9 ± 1012
rCF-PA3$_{60}$	3454.4	4159.7 ± 1148
rCF-PA5$_{60}$	3465.8	4565.1 ± 667
rCF-PL3$_{70}$	3321.6	4808.0 ± 1270
rCF-PL5$_{70}$	3320.7	6327.6 ± 965
rCF-PA3$_{70}$	3370.8	4618.7 ± 2747
rCF-PA5$_{70}$	3508.3	6846.8 ± 2021

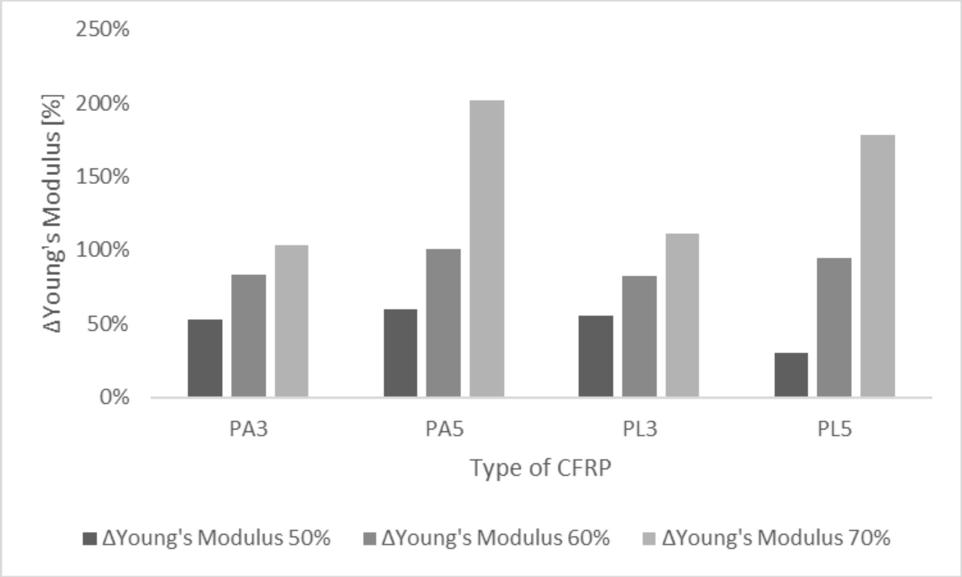

Figure 68: Improvement in Young's Modulus for each CFRP composed of hybrid yarns with 50%, 60%, and 70% recycled CF compared to tensile strength of neat resin

4.4 Conclusions

This Chapter focused on the analysis of unidirectional thermosetting composite materials reinforced with hybrid yarns composed of different percentages of recycled CF and thermoplastic fibres. Specifically, it was examined whether the characteristics of the hybrid yarns used for the CFRPs’ production affect their mechanical properties.

Overall, it was found that the amount of recycled CF and the number of draw frame doubling strongly impact on both tensile strength and Young’s Modulus values, while the thermoplastic fibre has no influence. The same may be applied for the 3-way interaction. Instead, different behaviours can be inferred for 2-way interactions for the two mechanical properties under consideration. In detail, in the case of tensile strength they are all statistically significant, while in the case of Young’s Modulus only the one between recycled CF and number of draw frame doubling is significant. In general, it is possible to state that the greater the amount of recycled CF as well as the number of draw frame doubling in the hybrid yarn, the greater the mechanical properties of the thermosetting CFRPs produced.

In the general conclusion, composites produced show better mechanical properties than the neat resin, thus highlighting that hybrid yarns produced are effective as reinforcement. Eventually, the best mechanical properties in terms of tensile strength and Young’s Modulus were measured in unidirectional thermosetting CFRP composites consisting of 70% recycled CF and 30% polyamide with a number of draw frame doubling equal to 5.

CHAPTER 5

Environmental sustainability of the innovative spinning process

5.1 Introduction

Environmental sustainability has been gaining momentum over the last years. Indeed, it has become a pivotal requirement for a product or a process owing to the growth of both legislative constraints and the environmental awareness of customers (Landi et al., 2022). Besides these reasons, the scarcity of energy resources and raw materials also plays an essential role (e.g. Girtan et al., 2021). Since the 1960s, LCA has been used to analyse solutions for sustainable productions (Buyle et al., 2013).

As already mentioned, CFs are a valuable raw material that deserves to be recovered to reduce environmental impact and recover embedded energy. In such a context, various LCA studies on CFRP recycling technologies have been conducted. Typically, they compare two or more recycling technologies, or one or more recycling technologies with landfilling and incineration to analyse the environmental impacts of decisions on the life cycle of a product from a holistic perspective (i.e. also considering end-of-life product decisions). For instance, Witik et al. (2013) compared pyrolysis to incineration and landfill disposal in terms of its environmental sustainability, while Prinçaud et al. (2014) investigated the environmental impact of solvolysis using supercritical water for recycling CFRP composites. Nevertheless, no LCA studies have been focused on the processes used for the manufacturing of semi-finished products or composites made from recycled CF, so far.

To bridge this gap, in this Chapter an LCA analysis of the developed innovative spinning process is presented. To the best of researcher’s knowledge, this is the first attempt to provide knowledge on the environmental sustainability of processes able to process recycled CFs to obtain semi-finished products with good specific properties that in turn can be used for value added applications. More in detail, in this study, the analysis is focused on the spinning process for the production of hybrid yarns composed of recycled

CFs from manufacturing scraps presented in Chapter 3 (Section 3.3).

The remainder of this Chapter is organised as follows. First, the Materials and methods Section defines the scope of the analysis and describes the Life Cycle. Then, the Life Cycle Impact Assessment is realized. Finally, LCA results are exposed with researcher' considerations in the Results and discussion.

5.2 Materials and methods

Since the LCA methodology (ISO 14044) is universally recognised as one of the most useful tools for quantitatively assessing the sustainability of current technologies, critically discussing the choices to implement during eco-design and evaluating the environmental performances of the new developed technologies, it was adopted to identify the environmental impacts of the proposed innovative spinning process (Hauschild et al., 2018).

Typically, two types of LCA are distinguished. One type of LCA aims to describe the pertinent in- and outflows from the life cycle along with its subsystems. This type of LCA is called attributional. The consequential LCA, on the other hand, aims to describe how the flows might change depending on the choices made (Finnveden and Potting, 2014). In this study, the life-cycle modelling adopted is of the attributional type (i.e. the technology and environmental models used are linear, stationary and they make use of the *Ceteris Paribus* assumption). In particular, what is not directly modified by the system at hand is not modelled (JRC, 2010).

According to ISO 14040 and ISO 14044, the LCA process consists of four phases: goal and scope definition, inventory analysis, impact assessment, and interpretation. These phases are better introduced in the following.

5.2.1 Goal and scope definition

This LCA study aims to assess the environmental impacts associated with the proposed innovative spinning process leading to the production of the hybrid yarns, identifying material and energy consumption hotspots, emissions to the environment and waste produced at each process step. Moreover, results' robustness was tested through a sensitivity analysis, which consists of evaluating the variation of environmental results following changes in relevant inputs.

The recycled CFs from manufacturing scraps were the raw materials and the ring-spun yarns were the end products. In detail, the hybrid yarn composed of 70% recycled CF and 30% polyamide with a number of draw frame doubling equal to 5 was selected as end product since it allows the production of the CFRP composite material showing the best mechanical properties, as documented in the previous chapter. Henceforth, this base case will be referred to as ‘Baseline’.

The system boundaries of the study, shown in Figure 69, were traced with a cradle-to-gate approach. This means that it refers only to a specific step of the life cycle of the ring-spun hybrid yarn. Specifically, all processes from the extraction of the raw materials to the production were considered, while the distribution, product use and disposal phases were not. The functional unit was set to 25 grams of material which corresponds approximately to 360 m of final product. This functional unit corresponds to the quantity of material actually used for the production and allows the comparison of different hybrid yarns independently of their final length.

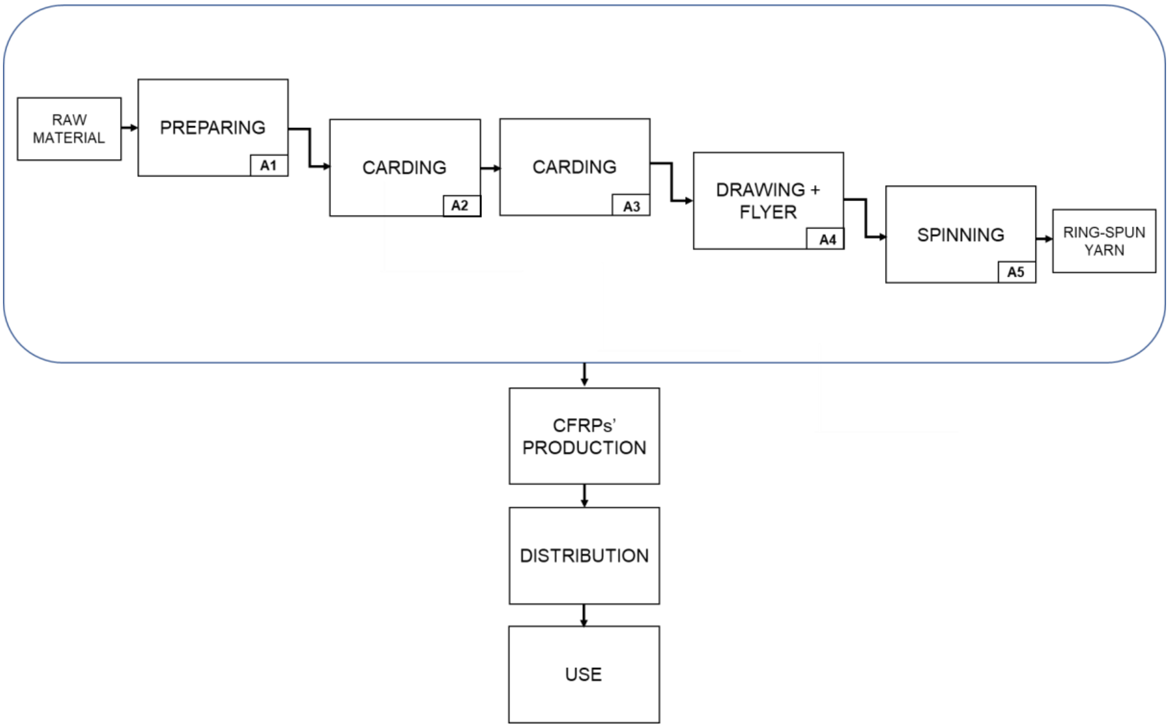

Figure 69: System boundary of the study. The carding phase is divided into A2 and A3 to represent the double passage described in Section 3.3

5.2.2 Life cycle inventory analysis

The fundamental procedure for evaluating the impact of the life cycle, allowing for the quantification of flows into and out of the system boundaries, is life cycle inventory. These flows encompass the use of resources (i.e. raw materials and energy), as well as releases into the air, water and soil related to the system. A list of the substances consumed and released into the environment and the amount of energy used was prepared during this stage. To this end, the production process was mapped according to the IDEF0 methodology, as it is a versatile modeling technique that accurately identifies the inputs, outputs, controls and mechanisms of the process activities (Bevilacqua et al., 2015). The result obtained is represented in Figure 70.

In this study, recycled CFs and polyamide fibres are the raw materials required for the production of ring-spun hybrid yarns using the innovative spinning process. The main materials, resources, energy consumption and waste produced in the process are shown in Table 29.

Table 29: Life Cycle Inventory Data

Phase	Flow	Quantity
Preparing (A1)	Recycled CF	17.5 g
	Polyamide	7.5 g
Carding (A2)	Electricity	0.15 kWh
	Waste fibre	1.8 g
Carding (A3)	Electricity	0.09 kWh
	Waste fibre	0.8 g
Drawing + Flyer (A4)	Electricity	0.06 kWh
	Waste fibre	0.5 g
Spinning (A5)	Electricity	0.18 kWh

Since this process is particularly innovative at laboratory level, according to ISO 14040, primary data (i.e. all materials and energy flows) for the process were collected in the laboratory facility. Instead, secondary data, such as those regarding extraction of resources and the production of materials and energies, were extrapolated from the updated literature and the international database Ecoinvent v.3.6 (Wernet et al., 2016) that is a comprehensive and reliable database which includes consistent and transparent Life cycle inventory data and methods commonly required for LCA research (Frischknecht and Rebitzer, 2005; Scrucca et al., 2020; Siracusa et al., 2014). As regards waste produced, it is essential to highlight that the one derived from the Preparing (A1) + Carding (A2), the Carding (A3) and the Spinning (A5) phases was considered to have zero impact and was therefore not included into the LCA analysis carried out. This is a reasonable choice as the scraps from Preparing (A1) + Carding (A2) and the Carding (A3) may potentially be reused for the production of compounds and therefore would not be disposed of in landfills. In the case of Spinning (A5), instead, the waste corresponds to a very limited amount of powder (about 0.04% of the total material) that is dispersed into the atmosphere and can therefore be disregarded. Eventually, the LCA study was modelled in SimaPro[5] v. 9.3.0.3 software.

5.2.3 Life cycle impact assessment

The life cycle impact assessment can be performed using a variety of methods. EF method 3.0 normalisation and weighting set - impact assessment method of Environmental Footprint initiative (Fazio et al., 2018), which has been proposed by the European Commission (European Commission, 2017), was the methodology employed in the current LCA analysis. It includes 16 impact categories, namely Climate Change; Ozone Depletion; Human Toxicity cancer; Human Toxicity non-cancer; Particulate matter disease incidence Ionising Radiation; Photochemical ozone formation; Acidification; Eutrophication terrestrial; Eutrophication freshwater; Eutrophication marine; Ecotoxicity freshwater; Land Use; Water use; Resource minerals and metals; and Resource use fossil. In order to avoid losing information defined by the European Commission, the researcher decided to not reduce the number of impact categories (Famiglietti et al., 2021)

5.3 Results and discussion

In this Section, the magnitude of potential environmental impacts of the innovative spinning process was determined and assessed through a dominance analysis. Additionally, to determine the impact of input data uncertainty on the outcomes and evaluate the robustness of assumptions and modelling options, both an uncertainty and sensitivity analysis were carried out (Andrianandraina et al., 2015).

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[5] https://simapro.com/

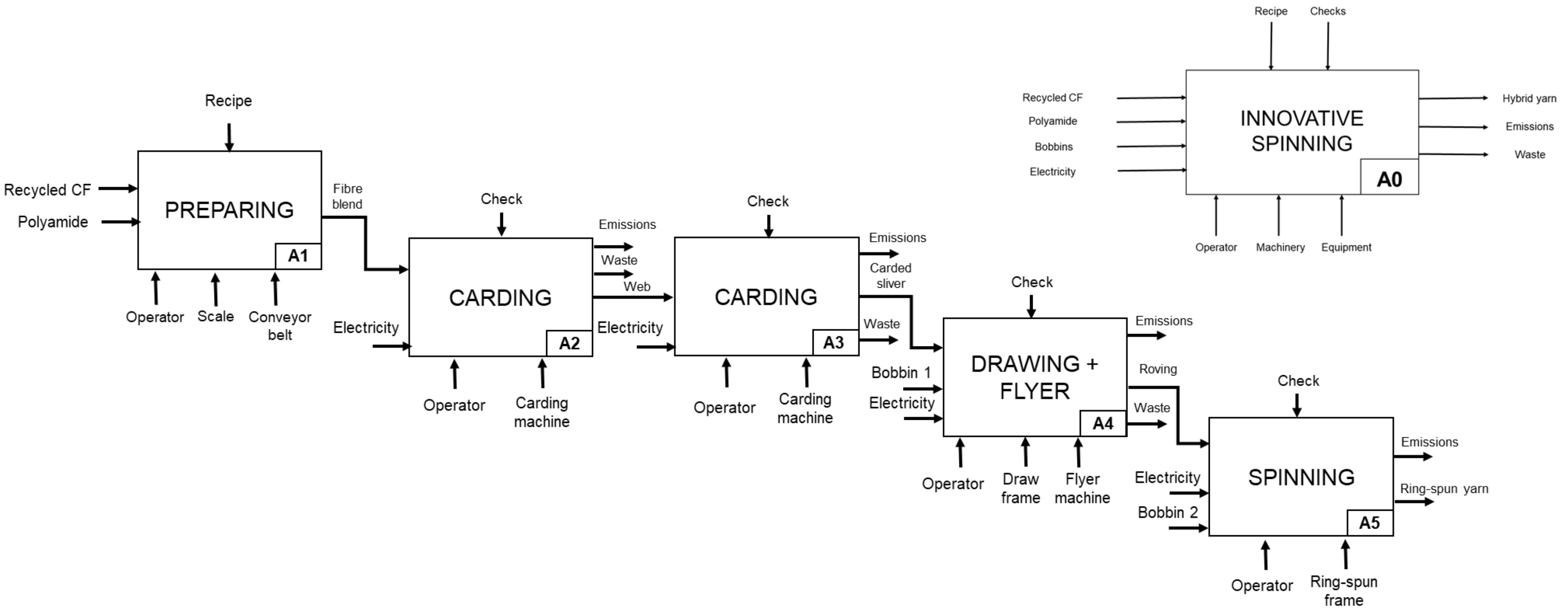

Figure 70: IDEF0 of the innovative spinning process

Table 30: Impact assessment findings for functional unit

Impact category	**Units**	**Characterisation**	**Units**	**Normalisation**	**Units**	**Weighting**	**%**
Climate change	kg CO_2 eq	2.64E-01	-	3,26E-05	µPt	6,86E+00	31.31%
Ozone depletion	kg CFC11 eq	2.57E-08	-	4,79E-07	µPt	3,02E-02	0.14%
Ionising radiation	kBq U-235 eq	2.57E-02	-	6,09E-06	µPt	3,05E-01	1.39%
Photochemical ozone formation	kg NMVOC eq	6.39E-04	-	1,57E-05	µPt	7,52E-01	3.43%
Particulate matter	disease inc.	6.87E-09	-	1,15E-05	µPt	1,03E+00	4.72%
Human toxicity, non-cancer	CTUh	2.02E-09	-	8,81E-06	µPt	1,62E-01	0.74%
Human toxicity, cancer	CTUh	7.64E-11	-	4,52E-06	µPt	9,63E-02	0.44%
Acidification	mol H+ eq	1.19E-03	-	2,14E-05	µPt	1,32E+00	6.05%
Eutrophication, freshwater	kg P eq	4.96E-05	-	3,08E-05	µPt	8,64E-01	3.94%
Eutrophication, marine	kg N eq	1.99E-04	-	1,02E-05	µPt	3,01E-01	1.37%
Eutrophication, terrestrial	mol N eq	2.13E-03	-	1,20E-05	µPt	4,46E-01	2.04%
Ecotoxicity, freshwater	CTUe	2.36E+00	-	5,54E-05	µPt	1,06E+00	4.85%
Land use	Pt	9.24E-01	-	1,13E-06	µPt	8,95E-02	0.41%
Water use	m^3 depriv.	1.49E-01	-	1,30E-05	µPt	1,11E+00	5.05%
Resource use, fossils	MJ	3.69E+00	-	5,68E-05	µPt	4,72E+00	21.55%
Resource use, minerals and metals	kg Sb eq	2.32E-06	-	3,65E-05	µPt	2,76E+00	12.58%

Table 31: Characterisation results for functional unit related to each phase of the innovative spinning process

Impact category	Units	Total	Preparing (A1) + Carding (A2)	Carding (A3)	Drawing + Flyer (A4)	Spinning (A5)
Climate change	kg CO_2 eq	2.64E-01	1.32E-01	3.60E-02	2.44E-02	7.09E-02
Ozone depletion	kg CFC11 eq	2.57E-08	8.01E-09	4.87E-09	3.22E-09	9.58E-09
Ionising radiation	kBq U-235 eq	2.57E-02	7.97E-03	4.88E-03	3.23E-03	9.61E-03
Photochemical ozone formation	kg NMVOC eq	6.39E-04	3.45E-04	8.09E-05	5.37E-05	1.59E-04
Particulate matter	disease inc.	6.87E-09	4.25E-09	7.18E-10	4.85E-10	1.41E-09
Human toxicity, non-cancer	CTUh	2.02E-09	7.11E-10	3.61E-10	2.41E-10	7.10E-10
Human toxicity, cancer	CTUh	7.64E-11	2.79E-11	1.33E-11	9.00E-12	2.62E-11
Acidification	mol H+ eq	1.19E-03	5.55E-04	1.74E-04	1.15E-04	3.43E-04
Eutrophication, freshwater	kg P eq	4.96E-05	1.64E-05	9.13E-06	6.04E-06	1.80E-05
Eutrophication, marine	kg N eq	1.99E-04	1.05E-04	2.57E-05	1.77E-05	5.06E-05
Eutrophication, terrestrial	mol N eq	2.13E-03	1.08E-03	2.86E-04	1.90E-04	5.64E-04
Ecotoxicity, freshwater	CTUe	2.36E+00	7.63E-01	4.40E-01	2.93E-01	8.66E-01
Land use	Pt	9.24E-01	2.88E-01	1.75E-01	1.16E-01	3.45E-01
Water use	m^3 depriv.	1.49E-01	6.15E-02	2.42E-02	1.60E-02	4.76E-02
Resource use, fossils	MJ	3.69E+00	1.73E+00	5.41E-01	3.58E-01	1.07E+00
Resource use, minerals and metals	kg Sb eq	2.32E-06	1.06E-06	3.49E-07	2.31E-07	6.87E-07

Table 32: Characterisation results for functional unit taking into account only Climate change, Human toxicity and Ecotoxicity freshwater

Impact category	**Units**	**Total**	**Preparing (A1) + Carding (A2)**	**Carding (A3)**	**Drawing + Flyer (A4)**	**Spinning (A5)**
Climate change - Fossil	kg CO2 eq	2.61E-01	1.32E-01	3.55E-02	2.41E-02	6.99E-02
Climate change - Biogenic	kg CO2 eq	2.70E-03	8.62E-04	5.05E-04	3.34E-04	9.94E-04
Climate change - Land use and LU change	kg CO2 eq	3.34E-05	1.05E-05	6.31E-06	4.18E-06	1.24E-05
Human toxicity, non-cancer - organics	CTUh	9.89E-11	4.80E-11	1.40E-11	9.31E-12	2.75E-11
Human toxicity, non-cancer - inorganics	CTUh	3.46E-10	1.64E-10	4.99E-11	3.34E-11	9.81E-11
Human toxicity, non-cancer - metals	CTUh	1.62E-09	5.23E-10	3.01E-10	2.01E-10	5.93E-10
Human toxicity, cancer - organics	CTUh	2.45E-11	7.65E-12	4.61E-12	3.17E-12	9.07E-12
Human toxicity, cancer - inorganics	CTUh	0	0	0	0	0
Human toxicity, cancer - metals	CTUh	5.19E-11	2.02E-11	8.73E-12	5.83E-12	1.72E-11
Ecotoxicity, freshwater - organics	CTUe	2.05E-02	8.20E-03	3.37E-03	2.24E-03	6.64E-03
Ecotoxicity, freshwater - inorganics	CTUe	2.07E-01	8.02E-02	3.45E-02	2.39E-02	6.80E-02
Ecotoxicity, freshwater - metals	CTUe	2.14E+00	6.74E-01	4.02E-01	2.67E-01	7.92E-01

5.3.1 Dominance analysis of the baseline

The dominance analysis aims at examining the life cycle stages that have the greatest impact on the environment (Baumann and Tillman, 2004). Table 30 summarizes the impact of each category in terms of absolute value, as well as after normalisation and weighting.

Once the weighting procedure has been carried out, the effect of Climate Change, Resource Use Fossils and Resource use, minerals and metals cumulatively contributed to around 65.4% of the total environmental of the innovative spinning process. Each of the other categories contributes significantly less than 10% to the total impact.

More details on the share of the stages composing the innovative spinning process for each impact category are shown in Table 31. The categories Climate Change, Human Toxicity and Ecotoxicity freshwater were exploded into further subcategories. In detail, Climate Change was subdivided into fossil, biogenic and land use and land-use change contribution. While Human Toxicity Non-cancer, Human Toxicity cancer and Ecotoxicity freshwater, were subdivided into organic, inorganic and metal. Table 32 shows the results of the characterisation of these impact subcategories.

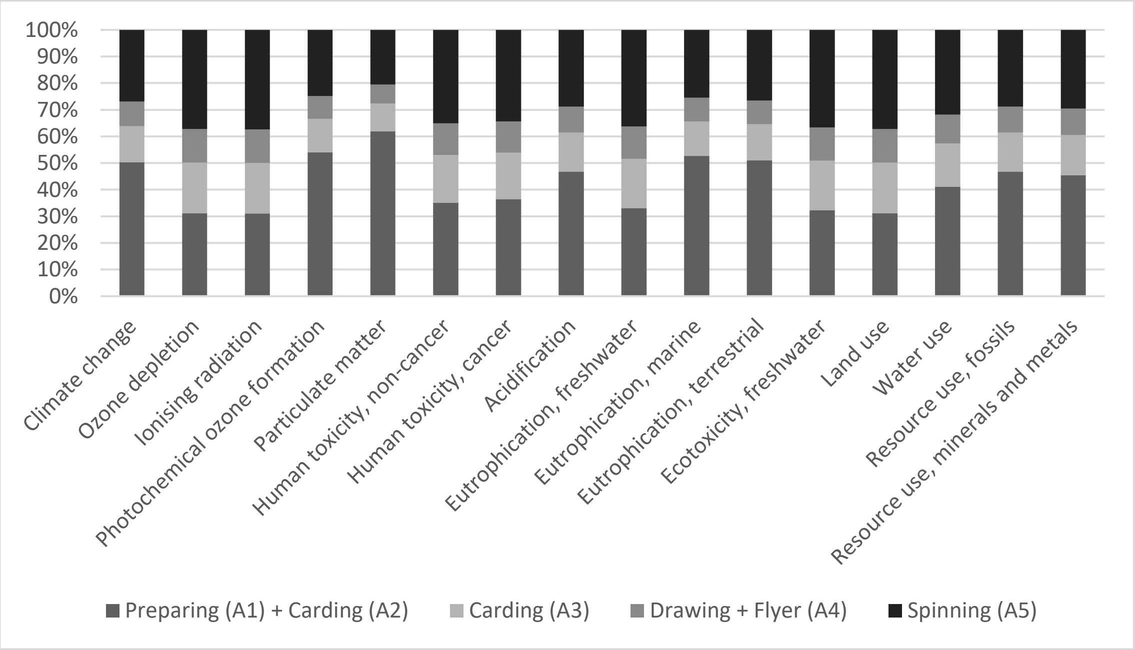

Figure 71: Contribution of the different phases of the innovative spinning process to each impact category

Figure 71 graphically shows the breakdown of the different impacts for the different phases of the process (whose specific values are also reported in Table 31). All in all, the main contribution to almost all the environmental impact categories concerns the Preparing (A1) + Carding (A2) phase. Indeed, it ranges from 61.9% for Particulate matter to 35.2% for Human Toxicity, non-cancer. In the other cases, the most impacting phase is Spinning (A5) ranging between 37.4% for Ionising radiation to 36.3% for Eutrophication, freshwater.

Since Preparing (A1) + Carding (A2) is the process' phase responsible for most of impact categories, it deserves further investigation. Specifically, it could be of interest the analysis of the share of its different input for each impact category. The results are depicted in Figure 72. Overall, it is possible to claim that the use of recycled CF from manufacturing scrap is not the main responsible for any impact category. Indeed, it accounts for at most 1% of the total value. On the contrary, the use of polyamide fibre strongly affects about half of the impact categories ranging from 72.6% (Particulate matter) to 55.8% (Climate Change). The same can be argued about electricity, which holds the highest values and ranges from 98.3% (Ionising radiation) to 53.0% (Resource use, minerals and metals). Acidification and Resource use, fossils are equally affected by the two inputs.

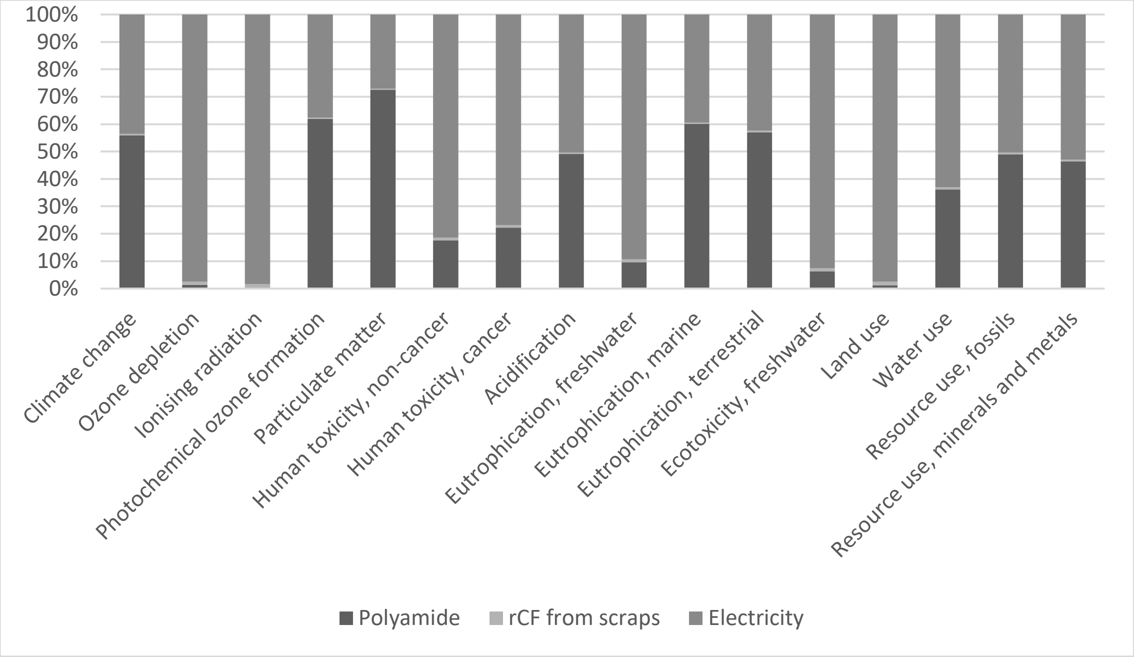

Figure 72: Details on the share of inputs of Preparing (A1) + Carding (A2) phase for each impact category

Besides, it could be interesting analysing the impact of the waste produced at the different stages of the process under consideration on the impact categories. Thus, considering the above, the only phase that leads to waste production turns out to be A4 (Drawing + Flyer). Table 33 reports the impacts of fibre waste (output) and electricity (input) of phase A4 to each impact category.

Table 33: Impact of waste and electricity belonging to Drawing + Flyer (A4) phase to each impact category

Protection Area	Waste	Electricity
Impact category	2.34%	97.66%
Climate change	0.10%	99.90%
Ozone depletion	0.03%	99.97%
Ionising radiation	0.46%	99.54%
Photochemical ozone formation	2.06%	97.94%
Particulate matter	0.99%	99.01%
Human toxicity, non-cancer	1.98%	98.02%
Human toxicity, cancer	0.17%	99.83%
Acidification	0.04%	99.96%
Eutrophication, freshwater	3.84%	96.16%
Eutrophication, marine	0.47%	99.53%
Eutrophication, terrestrial	0.57%	99.43%
Ecotoxicity, freshwater	0.22%	99.78%
Land use	0.18%	99.82%
Water use	0.07%	99.93%
Resource use, fossils	0.03%	99.97%

Overall, it is possible to state that it is electricity that almost completely impacts all impact categories. The highest contribution of waste, in fact, relates to the category Eutrophication, marine and is equal to 3.8%.

5.3.2 Uncertainty analysis

The uncertainty analysis was performed by means of the Monte Carlo method, using the specific calculation module included within the SimaPro v. 9.3.0.3 software. According to the Ecoinvent database, the Monte Carlo approach considers each input parameter as a stochastic variable with a lognormal probability distribution. In order to achieve convergence for both mean and variance values, the number of executions was set at 1000 with a 95% confidence interval (Raynolds et al., 1999) .

In this, Section the outcomes of the Monte Carlo Simulation are presented. In detail, Table

34 reports the results related to the different impact categories for the characterisation phase. Overall, barring a few specific categories, the level of uncertainty is fairly low.

Finally, the graph of the distribution obtained from the uncertainty analysis for the Climate Change category, the most impactful after normalisation (31.3%), and the most commonly analysed, is shown in Figure 73.

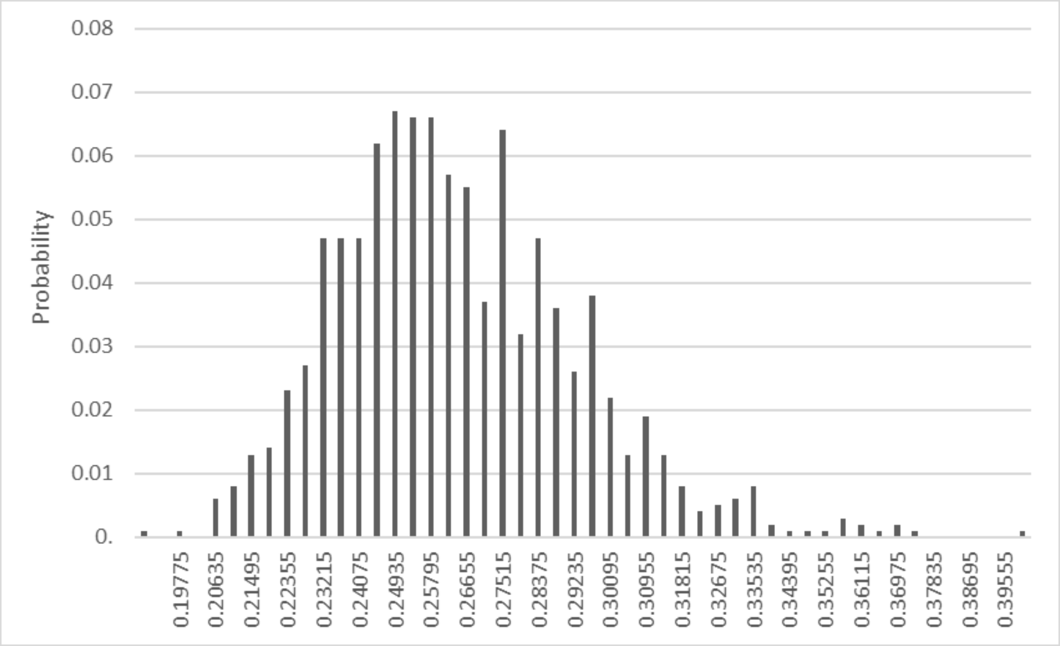

Figure 73: Distribution of the Climate Change impact category for characterisation results

5.3.3 Sensitivity analysis

A sensitivity analysis is aimed at estimating the effects of the choices made concerning methods and/or data of the study on the results. Generally, such analyses concentrate on the differences in outcomes associated to changes in major inputs (Andrianandraina et al., 2015).

In this research, the two following scenarios were considered:

- Polyamide fibre is replaced with polyester fibre (Scenario 1).
- Recycled CF from manufacturing scraps is replaced with recycled CF from pyrolysis (Scenario 2).

Table 34: Results of the uncertainty analysis through Monte Carlo method (SD: standard deviation; CV: Coefficient of variation; SEM: Standard error of mean)

Impact category	Unit	Mean	Median	SD	CV	2.50%	97.5%	SEM
Acidification	mol H+ eq	1,19E-03	1,18E-03	1,32E-04	1,11E+01	9,74E-04	1,48E-03	4,19E-06
Climate change	kg CO_2 eq	2,64E-01	2,60E-01	2,90E-02	1,10E+01	2,15E-01	3,31E-01	9,18E-04
Ecotoxicity, freshwater	CTUe	2,38E+00	2,28E+00	6,75E-01	2,84E+01	1,41E+00	3,97E+00	2,13E-02
Eutrophication, freshwater	kg P eq	4,97E-05	4,30E-05	2,85E-05	5,73E+01	1,88E-05	1,32E-04	9,01E-07
Eutrophication, marine	kg N eq	1,99E-04	1,96E-04	2,37E-05	1,19E+01	1,61E-04	2,50E-04	7,48E-07
Eutrophication, terrestrial	mol N eq	2,12E-03	2,10E-03	2,57E-04	1,21E+01	1,72E-03	2,67E-03	8,13E-06
Human toxicity, cancer	CTUh	7,84E-11	7,68E-11	5,98E-11	7,62E+01	-3,65E-11	2,03E-10	1,89E-12
Human toxicity, non-cancer	CTUh	2,17E-09	2,19E-09	6,92E-09	3,20E+02	-1,23E-08	1,59E-08	2,19E-10
Ionising radiation	kBq U-235 eq	2,64E-02	1,75E-02	2,66E-02	1,01E+02	7,91E-03	9,91E-02	8,41E-04
Land use	Pt	9,29E-01	9,09E-01	1,70E-01	1,83E+01	6,43E-01	1,31E+00	5,37E-03
Ozone depletion	kg CFC11 eq	2,59E-08	2,47E-08	7,00E-09	2,70E+01	1,56E-08	4,27E-08	2,21E-10
Particulate matter	disease inc.	6,89E-09	6,81E-09	6,13E-10	8,89E+00	5,91E-09	8,20E-09	1,94E-11
Photochemical ozone formation	kg NMVOC eq	6,39E-04	6,30E-04	7,05E-05	1,10E+01	5,27E-04	7,92E-04	2,23E-06
Resource use, fossils	MJ	3,72E+00	3,67E+00	5,41E-01	1,45E+01	2,82E+00	4,90E+00	1,71E-02
Resource use, minerals and metals	kg Sb eq	2,34E-06	2,29E-06	4,09E-07	1,75E+01	1,72E-06	3,33E-06	1,29E-08
Water use	m^3 depriv.	1,10E-01	1,20E+00	1,21E+01	1,10E+04	-2,75E+01	1,94E+01	3,84E-01

The choice fell on these inputs for several reasons. First of all, the study suggests that the use of polyamide fibre strongly contributes to a plethora of impact categories. Moreover, polyester was effectively used for the production of ring-spun hybrid yarns, as already described in Chapter 3. Therefore, in doing so, it is possible to find out the environmental impact of a real scenario. Second, recycled CF from pyrolysis is another important source of waste CF. Indeed, pyrolysis allows the recovery of CFs from end-of-life CFRPs (Pickering, 2006) which are currently increasing. Furthermore, it is a scenario that could potentially be applicable in the near future.

To model the production processes of the input materials, reference was made both to the background data of the available LCA database (i.e. EcoInvent) (Wernet et al., 2016) and to the scientific literature (e.g. Khalil, 2017).

Table 35 shows the characterisation results of LCA analyses performed, allowing the comparison between the Baseline, Scenario 1 and Scenario 2. Looking at the values, it is possible to draw some considerations. First, Baseline and Scenario 1 are quite similar to each other. Indeed, Scenario 1 shows good impact reductions (values range between -4.4% and -24.0%) for some impact categories, such as Particulate matter and Eutrophication, marine, but also important increases, such as in the Ozone depletion category where there is an increase of 451.5%. As regards Climate Change, one of the best known and most analysed categories, the use of polyester fibre leads to a reduction of 15.9%. Overall, it may be claimed that the environmental savings resulting from the adoption of one thermoplastic fibre over the other depend on the impact category under investigation.

Second, Scenario 2 is the worst in terms of environmental impact. Indeed, it is characterised by significant increases in impact values for all categories considered compared to Baseline and Scenario 1. As the contribution of impacts from steps A3, A4 and A5 of the innovative spinning process is unchanged in all scenarios and mainly driven by electricity consumption, this result may be attributed to recycled CF from pyrolysis and, in more detail, to its production process (i.e. thermal recycling via pyrolysis) that is energy intensive. In Figure 74 it is possible to observe that the contribution associated with recycled CF from pyrolysis is the most significant for all the impact categories under consideration.

Table 35: Results of LCA analyses - Comparison between Scenarios (Functional unit: 25 g of material)

Impact category	Unit	Baseline	Scenario 1	Scenario 2
Climate change	kg CO_2 eq	2.64E-01	2.22E-01	6.55E+00
Ozone depletion	kg CFC11 eq	2.57E-08	1.42E-07	7.41E-07
Ionising radiation	kBq U-235 eq	2.57E-02	2.78E-02	1.78E+00
Photochemical ozone formation	kg NMVOC eq	6.39E-04	5.67E-04	1.10E-02
Particulate matter	disease inc.	6.87E-09	5.22E-09	6.96E-08
Human toxicity, non-cancer	CTUh	2.02E-09	2.26E-09	3.48E-08
Human toxicity, cancer	CTUh	7.64E-11	8.85E-11	1.42E-09
Acidification	mol H+ eq	1.19E-03	1.05E-03	2.05E-02
Eutrophication, freshwater	kg P eq	4.96E-05	5.62E-05	3.01E-03
Eutrophication, marine	kg N eq	1.99E-04	1.66E-04	3.73E-03
Eutrophication, terrestrial	mol N eq	2.13E-03	1.78E-03	3.47E-02
Ecotoxicity, freshwater	CTUe	2.36E+00	2.85E+00	4.20E+01
Land use	Pt	9.24E-01	9.93E-01	1.18E+01
Water use	m^3 depriv.	1.49E-01	1.43E-01	3.49E+01
Resource use, fossils	MJ	3.69E+00	3.49E+00	1.14E+02
Resource use, minerals and metals	kg Sb eq	2.32E-06	2.12E-06	1.00E-05

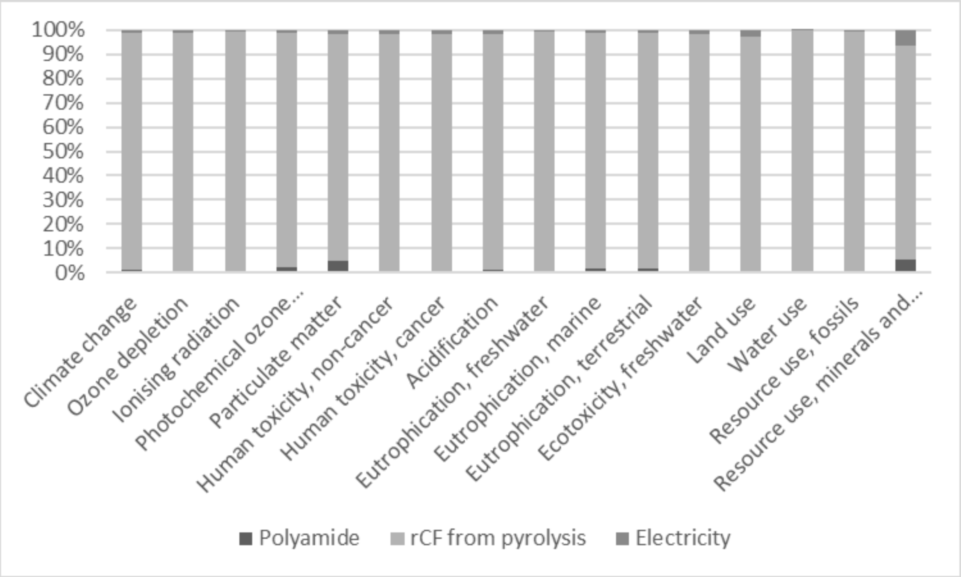

Figure 74: Details on the share of inputs of Preparing (A1) + Carding (A2) phase for each impact category - Scenario 2

5.3.3.1 Environmental Life Cycle Costing

To provide an idea of the cost associated to the LCA analysis performed, an Environmental Life Cycle Costing (eLCC) was carried out. eLCC is an LCA-based costing method where the functional unit, the scope and the system boundaries are the same (Hunkeler et al., 2008). Specifically, researcher adopted a steady-state modelling (i.e. without any temporal consideration), which assumes that all technologies remain constant over time. The Environmental Product Strategies (EPS) approach (Steen, 1999) which assesses the external costs of a product, a process or a service according to the willingness to pay to bring a deteriorated safeguard entity back up to a selected reference point (Afrane and Ntiamoah, 2012) was chosen. The environmental load unit (ELU), corresponding to 1 € cost, is the name given to this monetary amount.

In detail, The EPS 2015 dx method (Bengt, 2015) was used for the impact assessment. Its results are evaluated according to the willingness of an average OECD resident to pay to prevent environmental damage with reference to the state of the environment in the year 2015. Finally, the method assesses impacts from emissions and resource use that lead to significant changes in any of the following protection areas: Ecosystem Services, Access to Water, Biodiversity, Building Technology, Human Health and Abiotic Resources.

Table 36 reports the findings of the eLCC analysis concerning damage assessment. In accordance with the results of LCA analysis, the results of eLCC analysis show that Scenario 2 is the worst solution also in terms of environmental costs, while the Baseline, is the best one with a total value of 0.215 ELU per 1 functional unit. Anyway, such a value is very close to 0.236 ELU, value that characterises Scenario 1. Thus, confirming that the Baseline and Scenario 1 are quite similar to each other. Besides, it may be noticed that for all scenarios at hand, the damage categories making the largest contribution to the total value of environmental costs are Human health and Abiotic resources, which account for about 17.6% and 81.9% respectively in Baseline, about 14.7% and 84.5% in Scenario 1 and about 45.9% and 52.8% in Scenario 2.

Table 36: Results of eLCC analysis - Comparison between scenarios

Protection Area	Unit	Baseline	Scenario 1	Scenario 2
Ecosystem Services	ELU	1.02E-03	8.52E-04	2.54E-02
Access to Water	ELU	6.19E-05	5.19E-05	1.56E-03
Biodiversity	ELU	3.32E-06	2.80E-06	8.16E-05
Building Technology	ELU	8.80E-06	7.30E-06	2.29E-04
Human Health	ELU	3.78E-02	3.47E-02	9.97E-01
Abiotic Resources	ELU	1.76E-01	2.00E-01	1.15E+00
TOTAL	ELU	2.15E-01	2.36E-01	2.17E+00

5.4 Conclusions

In this Chapter, environmental impacts associated with the proposed innovative spinning process were assessed through the attributional life cycle modelling. In detail, an LCA and an eLCC analysis were performed following a cradle-to-gate approach, namely only all processes from the extraction of the raw materials to the production of ring-spun hybrid yarn were considered, and the functional unit was set to 25 grams of material. The Baseline considers the production of ring-spun hybrid yarns composed of 70% recycled CF from manufacturing scraps and 30% polyamide fibre.

The analysis showed that Climate Change, Resource Use Fossils and Resource use, minerals and metals cumulatively contribute to around 65.4% of the total environmental impact. Furthermore, the Preparing (A1) + Carding (A2) phase is the largest contributor to almost all environmental impact categories with values ranging between 61.9% and 31.2%. In the other cases, the most impacting phase is Spinning (A5) which ranges from

36.3% for Eutrophication, freshwater to 37.4% for Ionising radiation.

The uncertainty analysis carried out on the environmental impact results obtained for the Baseline showed that, with the exception of a few impact categories, the level of uncertainty is fairly low.

Furthermore, the sensitivity analysis exhibits that Scenario 2 (70% recycled CF from pyrolysis and 30% virgin polyamide) is the worst both in terms of environmental impact and externalities, while Baseline and Scenario 1 (70% recycled CF from manufacturing scraps and 30% virgin polyester) are quite comparable. Nevertheless, considering Climate Change impact category, this analysis highlights that environmental performance of the ring-spun hybrid yarn may be improved by replacing polyamide fibre with polyester. Indeed, there is a reduction of approximately 16% in the Carbon Footprint against an increase in environmental costs of 3%.

Overall, it may be concluded that the use of recycled CF from manufacturing scraps is preferable than the use of recycled CF from pyrolysis in terms of life cycle impacts. Moreover, since both solutions are characterized by similar environmental impact and their technical feasibility has been already tested, the choice between polyamide or polyester should rely on the best technical properties of the final CFRP composite.

CHAPTER 6

General conclusions

CFs are a valuable raw material from both a technical and an economic point of view. For this reason, they are increasingly being adopted in various sectors and, consequently, large quantities of CF waste will have to be managed in the near future. At the same time, sustainability and, more specifically, the circular economy are gaining momentum due to the increasing awareness of environmental issues. For these reasons, the establishment of technologies allowing the reuse of CFs in structural applications is essential. In such a context, the main goal of this Ph.D. project was to develop an innovative spinning process able to produce yarns made of recycled CFs with repeatable thermal-physical and mechanical properties and suitable for the production of good-quality CFRP composite materials.

In the first part of this thesis, where the first research question (*RQ1: Which type of CF waste source has the best properties?*) was addressed, a bibliometric analysis and a patent analysis were carried out to provide a comprehensive and holistic overview of the existing recycling technologies concurrently assessing the actual maturity level of each of them. Furthermore, hotspots and gaps, as well as future research directions were identified suggesting that more attention should be paid on remanufacturing processes.

Both analyses highlighted that the interest in the topic increased over the last decades likely due to the increased market knowledge of the sustainability issue and the pressure from the authorities. Nevertheless, scientific interest has been, and still is, different for each of the recycling macro-categories (for more details please refer to Chapter 2 of this manuscript). Besides these fundamental evidences, this part of the thesis also sheds light on the fibres' characteristics derived from each recycling technology.

In summary, chemical recycling provides fibres with high mechanical properties and length, but reduces the bonding capacity with the new resin and is generally not environmentally-friendly. Mechanical recycling overcomes this drawback while drastically decreasing the properties of the recycled fibres. A potential compromise is pyrolysis, which is an environmentally-friendly technology that allows fibres to be recycled with high mechanical properties depending on the process parameters. As an alternative, it has been widely recognised that recycled fibres from manufacturing scraps still present sizing and are therefore supposed to be easier to process. This information made it possible to understand where to concentrate the most practical efforts of the project. Indeed, focusing on designing an innovative spinning process on the basis of recycled CFs obtained by means of a technology which has little potential for future growth would likely not assure a solution maximising the potential of the recovered material.

The answer for the second research question (*RQ2: How can the traditional spinning process be adapted to deal with recycled CFs?*) was provided in the second part of the thesis, where the innovative spinning process developed was presented. Specifically, it emerged that recycled CFs from manufacturing scraps may be handled whether they are blended with a thermoplastic fibre and the amount of recycled CF able to provide hybrid yarns with good thermal-physical and mechanical properties ranges between 50% and 70% by weight. Such a result was achieved through a technical feasibility study consisting of two visual and two quantitative controls.

Ring-spun hybrid yarns' characterisation allowed the researcher to find out that, regardless of the type of thermoplastic fibre, the number of draw frame doubling seems to affect the tenacity of ring-spun hybrid yarns, while the increase in recycled CF content does not strongly affect the tenacity of ring-spun hybrid yarns. Nevertheless, the actual amount of remaining recycled CF, that is lower than that is actually inserted, seems to be correlated with the tensile strength value. Overall, ring-spun hybrid yarns composed of 70% recycled CF possess the best mechanical and thermal properties, regardless of the type of thermoplastic fibre.

From a more theoretical perspective, the high potential of ring-spun technology has been validated. Moreover, the obtained results proved that consolidated technologies can be exploited to foster circular economy after being appropriately revised. Therefore, in some cases, incremental technological innovations can be more fruitful than radical technological innovations.

In the third part, the analysis of unidirectional thermosetting composite materials reinforced with the ring-spun hybrid yarns produced in the previous step was reported. In particular, it was investigated whether the mechanical properties of the CFRPs were influenced by the features of the hybrid yarns employed in their manufacture. In this way, the third research question (*RQ3: What are the characteristics of the potential CFRPs produced by the innovative process?*) was answered.

By means of an ANOVA analysis, it emerged that the amount of recycled CF and the number of draw frame doubling strongly affect both tensile strength and Young’s Modulus values. On the contrary, the thermoplastic fibre has no influence as well as the 3-way interaction. As regards, the 2-way interactions, the greater the amount of recycled CF and the number of draw frame doubling in the ring-spun hybrid yarn, the greater the mechanical properties of the unidirectional thermosetting CFRPs produced.

Overall, it is possible to argue that composites manufactured generally exhibit better mechanical properties than the neat resin and, in detail, the ones composed of 70% recycled CF and 30% polyamide with a number of draw frame doubling equal to 5 appear to be characterised by the best mechanical properties in terms of tensile strength and Young’s Modulus.

From a more theoretical perspective, the possibility of producing good quality thermosetting unidirectional CFRPs from secondary raw material has been proven. Therefore, *ad hoc* applications can be purposely designed.

In the last part of this thesis, the last research question (*RQ4: What is the environmental impact of the developed spinning process?*) was addressed by investigating the environmental sustainability of the innovative spinning process developed. In detail, in order to assess the environmental impacts associated with the proposed innovative spinning process for the production of ring-spun hybrid yarns composed of 70% recycled CF and 30% polyamide fibre, an LCA analysis combined with an eLCC was performed on a 25 grams functional unit, following a cradle-to-gate approach.

Results showed that 65.4% of the total environmental impact is due to Climate Change, Resource Use Fossils and Resource use, minerals and metals, and almost all environmental impact categories are mainly impacted by the Preparing (A1) + Carding (A2) stage followed by Spinning (A5). Overall, the sensitivity analysis enabled to understand that the use of recycled CF from manufacturing scraps is preferable than the use of recycled CF from pyrolysis in terms of life cycle impacts and that the choice between polyamide or polyester should depend on the best mechanical properties of the final CFRP composite, as both solutions, which have already been tested in terms of technical feasibility, are characterised by fairly similar environmental impacts.

From a more theoretical perspective, this thesis provides the first work on the environmental sustainability of a technology that can process recycled CF to obtain semi-finished products with good mechanical and physical properties that in turn can be used for value-added applications.

Eventually, it is worth mentioning that the research carried out within this Ph.D. project supported a regionally funded project for the development of the innovative spinning process on an industrial scale.

6.1 Limitations and suggestions for further research

Of course, like any research project, this thesis does not come without limitations, which can be properly addressed in future research.

First, this Ph.D. project focused on the analysis and the use of recycled CF from manufacturing scraps for the production of ring-spun hybrid yarns. In order to expand the applicability of the proposed innovative spinning process, it could be useful to replicate the feasibility study and characterise the intermediate ring-spun hybrid yarn and the final composite material using another source of waste CF, such as recycled CF from pyrolysis. In this way, it would be possible to compare the results obtained and evaluate *ad hoc* applications.

Second, the ring-spun hybrid yarn twist was set at 300 tpm. This choice has negatively affected the tensile properties of the CFRPs produced, since it is well recognised that high mechanical properties in composites composed of discontinuous CF may be achieved through low twist yarns (Abdkader et al., 2022). Nevertheless, it was taken so that the electrical conductivity of the yarns could be enhanced. Indeed, in the near future research is planned on the production and characterisation of fabrics consisting of the manufactured hybrid yarns to assess their electrical properties.

Third, owing to the laboratory scale of the process and the limited quantity of ring-spun hybrid yarn produced, it was consciously decided to fabricate thermosetting CFRPs. Since it is, however, well known that thermoplastic matrix composites are more easily recyclable, a future development of this work will be the realisation of unidirectional thermoplastic CFRP starting from the obtained hybrid yarns. In this way, it would be possible to compare the results with what already exists in the scientific literature and thus potentially demonstrate the positive contribution of the ring-spinning phase. In the future, it would also be interesting to assess how and to what extent the results would change if recycled polymer fibres were used from a circular economy perspective.

Lastly, the LCA analysis was limited to the innovative spinning process in order to analyse the environmental sustainability of the process more closely resembling industrial spinning. Indeed, within this Ph.D. project, the stages of weaving the reinforcement and making the composite were carried out manually and would therefore not be representative of final industrial process in terms of the flows and procedures to be considered. In this regard, it could be of interest widening the system boundaries of the study to consider composites' production particularly in the case of thermoplastic CFRPs, especially following the industrialisation in a pilot plant of the process.

Moreover, assuming the same inputs and outputs, it would be interesting to carry out an LCA analysis to compare the proposed innovative spinning process with the one already found in the literature that uses other spinning methods, such as wrap or friction spinning (Akonda et al., 2012; M.M.B. Hasan et al., 2018).

To conclude, a *conventional LCC* should be performed to assess the pure economic feasibility of the proposed innovative spinning process, ultimately defining the economic incentive for companies to adopt such a process at an industrial scale (Hunkeler et al., 2008).

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