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Process development of pre-impregnated hybrid thin-ply composites

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par

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Alexios Argyropoulos

Abstract

Thin-ply composites, obtained with recently developed fiber spreading techniques, rapidly gained industrial interest because they offer a large composite design freedom, and lead to a composite tensile strength that is close to that of the individual fibers, as well as improved fatigue properties. However, their wider adoption towards critical lightweight composite applications remains limited due to their inherent toughness reduction and high production cost.

Fiber hybridization demonstrates a great potential for alleviating some of these drawbacks and promoting the manufacturing of laminates with balanced characteristics. Currently, most studies on thin ply hybrids employ simple interlayer configurations mainly due to difficulties in manufacturing more complex hybrid architectures, combining fibers that do not have similar diameters, stiffness, surface conditions and spreading characteristics. However, recent model predictions indicate that significant mechanical performance improvements should be attained with carefully designed intralayer (tow-by-tow) and intrayarn (fiber-by-fiber) hybrid architectures.

This research, performed at North Thin Ply Technology Sarl (NTPT) and EPFL in the framework of the European project Hyfisyn, aims to evaluate the relative influence of key process parameters on the quality, in terms of microstructural organization as well as resulting composite properties, of thin-ply hybrid prepregs and composite laminates. To reach this objective, the processing variables for fiber spreading and impregnation were analyzed, to adapt and improve the current pilot and industrial prepreg line aiming for the cost effective, yet high quality production of thin-ply composites prepregs at the ply-by-ply, tow-by-tow, and fiber-by-fiber levels.

A first analysis was carried out on thin-ply glass fiber prepregs, to evaluate the effect of mechanical pre-spreading and thermal treatment on the resulting single fiber and prepreg mechanical properties. The statistical strength distribution of the fibers and resulting composite strength was indeed negatively affected by the treatments, although fiber and composite modulus remained unchanged, opening some possibilities to modify the current process with limited strength reduction.

Then, tow level hybrid prepregs were progressively optimized, first using glass and carbon fibers, then two types of carbon fibers with high and low strain to failure, until reaching high quality prepregs. The next step was to produce intimately commingled hybrid prepregs. However, the manufacturing challenges encountered in the controlled mixing of dissimilar fibers for the production of hybrid thin-ply prepregs at an industrial scale made this process too unreliable. As an alternative, a novel calendering method, for fiber-level hybrid prepreg tape manufacturing was developed and showed promising results.

Composites were produced with several combinations of tow by tow and fiber by fiber hybrid configurations. New tools were developed to analyze the microstructure and quantify the degree of hybridization of these carbon fiber hybrids, highlighting the high quality of the materials obtained. Unnotched tensile tests were carried out, showing that the composite stiffness and strength did not reveal any additional fiber damage due to the processes.

The study contributed to development of thin ply hybrid composites, with hybridization degrees that reached unprecedented levels, and pointed the practical limitations to reach a high degree of commingling with industrial relevance in mind.

Keywords

Fiber reinforced polymers, Thin-ply composites, Composite manufacturing processes, Fiber-hybrids, Microstructural analysis.

Résumé

Les composites à plis fins, obtenus grâce à des techniques d'étalement des mèches de fibres récemment développées, ont rapidement suscité l'intérêt de l'industrie car ils offrent une grande liberté de conception des composites et permettent d'obtenir une résistance à la rupture en traction proche de celle des fibres individuelles, ainsi que de bonnes propriétés de résistance à la fatigue. Cependant, leur adoption plus large pour des applications critiques de composites légers reste limitée en raison de leur réduction inhérente de la ténacité et de leur coût de production élevé.

L'hybridation des fibres présente un grand potentiel pour atténuer certains de ces inconvénients et promouvoir la fabrication de stratifiés aux caractéristiques équilibrées. Actuellement, la plupart des études sur les composites à plis fins hybrides utilisent des configurations intercalaires simples, principalement en raison des difficultés de fabrication d'architectures hybrides plus complexes, qui doivent combiner des fibres qui n'ont pas le même diamètre, la même rigidité, les mêmes conditions de surface et les mêmes caractéristiques d'étalement. Cependant, les modèles récents indiquent que des améliorations significatives des performances mécaniques devraient être obtenues avec des architectures hybrides dispersées à l'intérieur des couches (mèche par mèche) et à l'intérieur des plis (de fibre en fibre).

Cette recherche, menée à North Thin Ply Technology Sarl (NTPT) et à l'EPFL dans le cadre du projet européen Hyfisyn, vise à évaluer l'influence relative des paramètres clés de la mise en œuvre des pré-imprégnés plis fins hybrides sur la qualité, en termes d'organisation microstructurale et de propriétés mécaniques résultantes, des stratifiés composites. Pour atteindre cet objectif, les paramètres clé pour l'étalement des fibres et l'imprégnation ont été analysés, afin d'adapter et d'améliorer les lignes pilote et industrielle actuelles visant une production rentable et de haute qualité de composites préimprégnés plis fins hybridés aux échelles des plis, mèches ou fibres.

Une première analyse a été effectuée sur des fibres de verre préimprégnées à plis fins, afin d'évaluer l'effet du pré-étalement mécanique et du traitement thermique sur les propriétés mécaniques de la fibre unitaire et du composite. La distribution statistique de la résistance des fibres et la résistance du composite résultant ont en effet été affectées négativement par les traitements, bien que le module des fibres et du composite soit resté inchangé, ce qui ouvre des possibilités de modifier le processus actuel avec une réduction limitée de la résistance.

Ensuite, les préimprégnés hybrides mèche par mèche ont été progressivement optimisés, d'abord en utilisant des fibres de verre et de carbone, puis deux types de fibres de carbone avec une déformation à la rupture élevée et faible, jusqu'à obtenir des préimprégnés de haute qualité. L'étape suivante consistait à produire des préimprégnés hybrides intimement mélangés. Cependant, les difficultés de fabrication rencontrées pour mélanger intimement les fibres dissemblables pour la production de préimprégnés hybrides à l'échelle industrielle ont rendu ce processus trop peu fiable. Une nouvelle méthode de calandrage pour la fabrication de bandes préimprégnées hybrides au niveau des fibres a été mise au point et a donné des résultats prometteurs.

Des composites ont été produits avec plusieurs combinaisons de configurations hybrides mèche par mèche et fibre par fibre. De nouveaux outils ont été développés pour analyser la microstructure et quantifier le degré d'hybridation de ces hybrides de fibres de carbone, soulignant la haute qualité des matériaux obtenus. Des essais de traction ont été réalisés, montrant que la rigidité et la résistance des composites ne révélaient aucun dommage supplémentaire des fibres dû aux processus.

L'étude a contribué au développement de composites hybrides à couches minces, avec des degrés d'hybridation qui ont atteint des niveaux sans précédent, et a mis en évidence les limites pratiques pour atteindre un degré élevé d'hybridation tout en gardant à l'esprit la pertinence industrielle.

Mots-clés

Matériaux composites, composites à plis fins, Procédés de mise en oeuvre, hybridisation, analyse microstructurale.

Contents

List of Figures

List of Tables

Chapter 1 Introduction

1.1 Motivation

The motivation behind this thesis stems from the broader context of addressing climate change and reducing greenhouse gas emissions, as outlined in the Marie Skłodowska-Curie Actions (MSCA) Innovative Training Network (ITN) H2020-MSCA-ITN-2014 funding this project. The domestic transport greenhouse gas (GHG) emissions increased in the European Union (EU) steadily between 2013 and 2019, while the 13.6 % decrease in emissions between 2019 and 2020 is attributed to the drastic decrease in all transport activities during the covid-19 pandemic was temporary. Road transport constitutes the higher proportion of overall transport emissions. In 2020 road transport emitted 77% of all EU's transport GHGs.

An irreversible shift to low-emission mobility has been proposed by the EU as a solution to this major environmental issue. By 2050, greenhouse gas emissions from transport will need to be at least 60% lower than in 1990 and on the path towards zero. However, the direct link between the weight of vehicles and energy consumption is an obstacle for meeting the emission targets of the transport sector with currently available technologies [1]. Apart from EU's environmental regulations, consumers' demand for fuel-efficient vehicles also encourages manufactures to invest in research and development of lightweight materials, and to evaluate the potential impact on the overall life cycle of the vehicle to ensure that potential increase in environmental impact at the production phase is compensated by fuel efficiency over the use phase of the vehicle [2] [3] [4] [5].

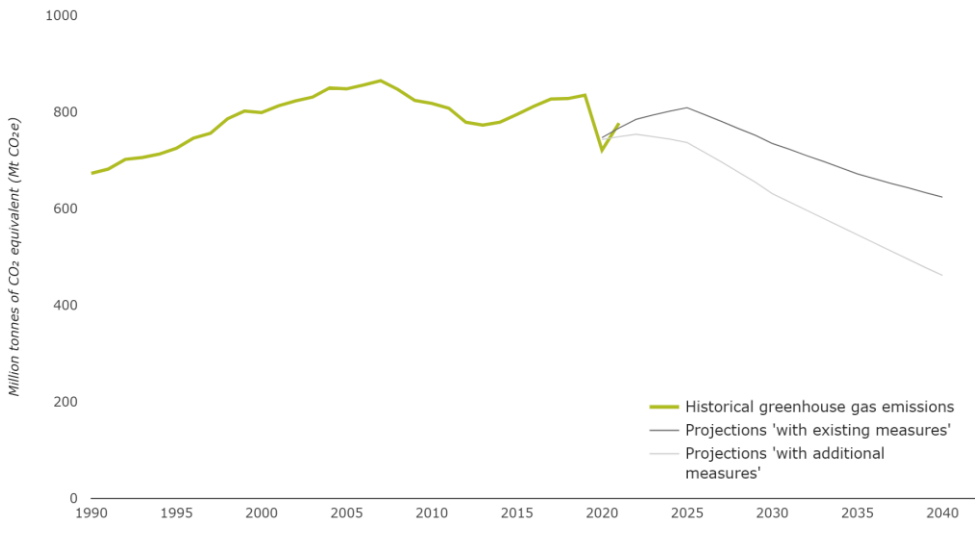


Figure 1.1 Greenhouse gas emissions from transport in Europe [1].

Fiber-reinforced polymers (FRPs) exhibit outstanding mechanical properties together with low density, which make them highly desirable in applications where weight reduction is crucial [6]. Among FRPs, fiber-hybrid composites, consisting of two or more fiber types in a common matrix have emerged as a promising solution to address various challenges in the field of composite materials, by adding one more possibility to tailor stiffness and strength, and possibly damping, electrical conductivity or other functional properties [7]. Moreover, when carefully designed, they have the potential to achieve synergetic effects, where the combined performance of the hybrid composite exceeds that of its individual constituent, bringing improvements in terms of mechanical performance, weight reduction, and cost efficiency that are highly desirable from the composites industry.

This study is part of a research and training project: HyFiSyn (Hybrid Fiber-reinforced composites: achieving Synergetic effects through microstructural design and advanced simulation tools), funded by the European Union’s Horizon 2020 research and innovation program under the Marie Sklodowska Curie grant agreement No 765881. HyFiSyn aims to extend the current understanding in the field of composite materials and address key cost, performance, recyclability, and manufacturing challenges faced by the composite industry through fiber hybridization.

More specifically, the critical importance of microstructure in achieving synergetic effects has been acknowledged by research on simulation tools that can predict the optimal fiber hybrid architecture for performance enhancements. However, the ability to validate these predictions has been limited since the current composite manufacturing processes have insufficient hybridization capability, due to a lack of understanding of the mechanisms that would enable the controlled mixing of dissimilar fibers at the desired ratios, thicknesses, and hybrid configurations.

This research thus focuses on the development and optimization of innovative manufacturing techniques at an industrial scale, to achieve the desired microstructural control in fiber hybrid composites. The shift of the manufacturing-microstructure paradigm to microstructure-manufacturing is one of the primary motivations of the HyFiSyn framework. This paradigm shift involves deliberately controlling the manufacturing process to yield target microstructures, rather than adjusting the microstructure to match the manufacturing process. Such an approach is critical for maximizing the potential benefits of fiber-hybrid composites and composites materials in general.

With this goal in mind, this research was conducted in an industrial setting, at the company North Thin Ply Technology Sarl (NTPT) in partnership with the Laboratory for Processing of Advanced Composites (LPAC) at the Swiss Federal Institute of Technology Lausanne (École Polytechnique Fédérale de Lausanne – EPFL). North Thin Ply Technology is a Swiss-based company that specializes in the field of advanced composite materials. Founded in 2005, NTPT has established itself as a world leader in the production of ultra-thin, high-performance composite materials using its proprietary fiber spreading and impregnation technology. One of

NTPT's most remarkable products is its thin-ply prepreg, which features carbon fiber plies with a thickness down to 15 microns. This innovative product is known for its high strength-to-weight ratio and consistent quality, making it well-suited for use in demanding applications where weight savings are crucial, such as in aerospace, motorsports, and sailing but also luxury goods due to its unique aesthetics [8].

1.2 Objectives

To propose scientific innovation and development in the processing of pre-impregnated hybrid thin-ply composites with complex hybrid architectures at an industrial scale, this work focuses on several objectives:

- Analyze the processing variables in state-of-the-art fiber spreading and impregnation techniques that could lead to best-performing hybrid prepregs.
- Develop and adapt the production equipment to achieve high-quality prepregs at low areal density for applications in hybrid composites, and to produce hybrid thin-ply composites prepregs at the ply-by-ply, tow-by-tow, and fiber-by-fiber levels.
- Investigate the potential effects of the fiber spreading techniques on the mechanical properties of the fiber, and of the resulting composite.
- Produce composite plates with the developed hybrid prepregs, analyze their overall quality, commingling degree using microstructural descriptors, and mechanical properties.
- Develop and supply various configurations of hybrid thin-ply prepregs for other researchers of the HyFiSyn consortium.

The main research questions are as follows:

- What is the optimal fiber hybridization level and related processing route to reach the desired mechanical properties, while preserving cost-efficiency?
- What is the added potential of manufacturing composites made from unidirectional pre-impregnated tapes of technical fibers that combine the low thickness (thin-ply) and fiber co-mingling (hybrid) effects?

The study contributes to the understanding of the effects that different manufacturing routes have on the overall performance of thin-ply composites through the analysis of their microstructural features and mechanical performance.

1.3 Confidentiality Agreement

Due to the nature of this industrial PhD program and the involvement of North Thin Ply Technology (NTPT), certain aspects of the research presented in this thesis are subject to confidentiality agreement between all parties involved (the researchers, NTPT, and EPFL) to protect the company's intellectual property.

While every effort has been made to provide as much detail as possible within the bounds of the agreement, it may not be possible to fully replicate certain aspects of the research without direct access to the confidential materials and processes of NTPT. We acknowledge that academic research often prioritizes reproducibility of results, but in the context of industrial PhD programs that involve confidential materials and processes, this may not always be possible. Nonetheless, the research presented in this thesis was conducted rigorously and the results presented are valid within the bounds of the agreement.

1.4 Thesis outline

This doctoral thesis is organized into 7 chapters, including the introduction and the conclusions. Each chapter addresses different aspects of thin-ply prepreg manufacturing, composite processing, and testing that are directly connected to the overall objective of developing and evaluating of new hybrid thin-ply prepregs and composites manufacturing methods.

After the introduction (**Chapter 1**), which provides an overview of the research background, problem statement, and research motivation and objectives, the thesis is organized according to the following structure:

Chapter 2 presents a systematic literature review of fiber-reinforced composite materials, with a particular focus on prepreg technology and state-of-the-art fiber spreading and impregnation methods that have contributed to the recent advancement of thin-ply prepreg and thin-ply composite material manufacturing. Additionally, this chapter introduces fiber hybridization as a promising strategy for alleviating the current major drawback of composite materials and highlights the key manufacturing challenges for achieving thin-ply composites with complex architectures. Chapter 2 also introduces the current microstructural analysis techniques as characterization methods for evaluating the level of fiber co-mingling in hybrid composite materials. Finally, the chapter concludes with a critical analysis of the current state-of-the-art thin-ply prepreg manufacturing and hybrid composites, highlighting the limitations and areas of improvement that will come by the development of processing methods for the fabrications of hybrid-thin ply prepregs with controlled characteristics.

Chapter 3 focuses on the materials and methods and describes in detail all the experimental procedures and techniques used to carry out the research. The chapter is divided into two parts: the materials used in the study and the methods used to perform the experiments. In the materials section, a detailed presentation of all the characteristics of the fibers and the resin system used for the fabrication of the prepregs is provided. The materials section then continues with the characteristics of the prepregs produced with each of the manufacturing methods that were tested in this study. In the method section, all the experimental procedures used in the study are explained in detail, with a focus on the optimization of existing procedures and the development of new procedures that allowed us to address manufacturing and characterization issues introduced by fiber hybridization. The method section also presents the microstructural analysis techniques and software tools that enabled the extraction of statistical data from hybrid composite cross-sections, which was the main tool for studying all the prepreg manufacturing and composite processing methods developed in this work. The selection of fibers that promote synergetic effects, the use of resin systems for improved toughness, and the examination of optimal stacking configurations for the manufacturing of multi-layer thin plies will be the main strategies applied during my project.

Chapter 4 describes the non-confidential stages of the North Thin Ply Technology (NTPT) fiber spreading and impregnation line that was employed for the manufacturing of non-hybrid/single fiber thin-ply prepregs to provide a general overview of the key equipment components and their role. The chapter also introduces an experimental thin-ply prepreg line that was based on existing equipment and allowed for the study of the effects of different processing routes on the quality of thin-ply prepregs. Finally, the chapter presents characteristic non-hybrid single-fiber thin-ply prepregs that were produced for this thesis, their microstructural profile, and baseline mechanical properties. The objectives of this chapter were to gain a deeper understanding of how processing steps may affect the quality of the fibers and, consequently, the quality of the final product. Additionally, this chapter aimed to study the existing prepreg production methods of NTPT and gain a deeper understanding of the mechanisms that govern fiber spreading and impregnation at an industrial level by monitoring the influence of key production parameters on the overall quality of single fiber thin-ply prepregs. Lastly, this chapter investigates the effects of the manufacturing process on the quality of the composites.

Chapter 5 presents the processing steps that enabled the industrial production of tow-level hybrid prepregs and the role of newly developed equipment stages in the controlled manufacturing of tow-level hybrids. The chapter describes the types of materials produced for this study and evaluates their microstructural profile to ensure that this level of hybridization can be achieved without any quality compromises. The objective of this chapter is to highlight the key processing steps and equipment required to produce tow-level hybrid prepregs.

Chapter 6 begins by presenting the hybridization attempts with the simultaneous fiber spreading approach that were conducted using the modified North Thin Ply Technology line to produce fiber-level hybrid prepregs. The chapter explores the potential of hybridization at a fiber level with the modified line and describes why this method did not produce the desired level of controllability and hybridization. The chapter then addresses the key manufacturing challenges encountered in the controlled mixing of dissimilar fibers for the production of hybrid thin-ply prepregs at an industrial scale. It proposes the calendering method, a novel manufacturing method for fiber-level hybrid prepreg tape manufacturing, and describes the main principle of the method. The objective of this chapter is to evaluate the potential of different manufacturing methods for producing fiber-level hybrid prepregs and to study their effects on the quality of the resulting composites.

Chapter 7 Presents key results of the analysis of the microstructural and the mechanical performance of the hybrid thin-ply prepregs and composite materials that were developed in the preceding chapters. Several types of configurations are explored, based on tow by tow and fiber level hybrid materials, as compared to baseline thin ply reference composites.

Chapter 8 Presents the key results and summarizes the primary conclusions of this study, along with a discussion of limitations and suggestions for future work.

Chapter 2 State of the art

2.1 Introduction to composite materials

Composite materials are a class of materials formed by combining two or more distinct constituent materials, each with different physical and chemical properties, to create a new synergistic material that exhibits unique combinations of properties. Of particular interest for modern engineering are the composites that are fabricated with multiple layers of technical fibers or textiles such as carbon, glass, and aramid that are held together, with a polymer matrix that acts as a binder [9].

Composite materials offer numerous advantages over traditional materials, making them increasingly adopted in various manufacturing industries. These advantages include weight reduction, corrosion resistance, part-count reduction, electromagnetic transparency, toughening for impact, erosion and wear resistance, acoustic and vibration damping, enhanced fatigue life, thermal/acoustical insulation, low thermal expansion, low or high thermal conductivity, self-healing, low or high permeability, fire resistance and fire retardancy, ablation, protection from lightning strikes, magnetoelectric response.

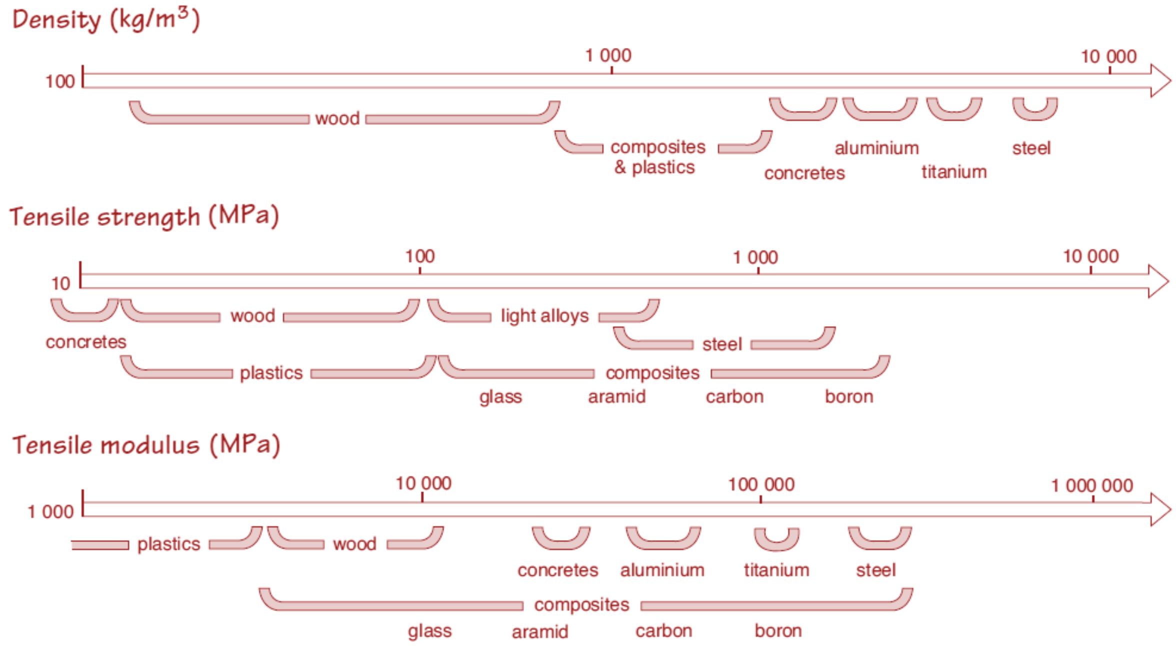


Figure 2.1 Comparison of characteristics of traditional and composite materials highlighting that composites can provide the advantages of lower weight, greater strength, and higher stiffness [10].

In composite materials, the reinforcing fibers are considered the primary load-bearing component as they are responsible for providing most of the composite's tensile strength, stiffness, and fatigue resistance. Resin is often considered the weaker constituent due to its relatively low tensile strength and stiffness compared to fibers. However, apart from its ability to provide adhesion, the matrix resin also helps to maintain fiber alignment and the overall structural integrity of the composite structure. It also serves to transfer loads between the fibers and improve the composite's resistance to compression and impact loads. The compressive strength of a composite is often only limited by the strength of the matrix [11]. Additionally, polymer matrices play a critical role in the protection of the composite against environmental factors such as moisture, UV radiation, and chemical attacks. Reinforcing fibers are susceptible to degradation if they are directly exposed to the environment. The resin provides a sealing that therefore is crucial for the corrosion resistance of the composite.

From a composite manufacturer's perspective, the resin matrix also determines the handling and processing characteristics of the intermediate and final production stages of a composite material as most composite fabrication methods are largely dependent on the resin processing requirements. Thermoplastic matrices, which consist of polymer materials that can be melted and solidified repeatedly through the application of heat without significant chemical changes, are typically heated, melted, and injected or pressed into a mold, where they solidify together with the reinforcements upon cooling to determine the final component geometry. Thermosetting resins, on the other hand, consist of polymer materials that undergo an irreversible chemical reaction known as curing or crosslinking, during the composite manufacturing process that transforms the resin from a liquid or semi-liquid state to a solid, three-dimensional network structure and ensures superior performance in specific applications. While techniques like hand lay-up, resin transfer molding, and filament winding have their merits, this research places a particular emphasis on prepregs composite manufacturing dues to their unique advantages for high-end composite applications[12].

2.2 Prepreg technology

Prepreg is a common technical term for reinforcing fibers such as carbon, glass, and aramid or even natural fibers such as flax which have been pre-impregnated with a specially formulated resin matrix system that usually includes all the proper curing agents. Prepregs are typically manufactured in unidirectional (UD) or fabric format and are delivered ready to use for lamination and molding without requiring additional resin or hardeners [13] [14] .

The real advantage of prepregs is that they feature a controlled ratio of resin to fabric which is difficult to achieve with traditional hand layups. Typical hand laminates even when vacuum bagged, can end up with excess resin which compromises the strength of the laminate. By yielding perfect resin content, prepregs pave the way to increased strength, part uniformity, and repeatability with less mass, less waste, and less curing time. The convenience of prepregs does come at a higher price though. Ultimately, they are more expensive than the dry fabric counterparts which factor in the individual cost of fabric, resin, and hardener that traditional hand layup methods require, and must be stored at controlled temperature, to avoid premature cure. So, they are mostly used for applications where the best balance of properties is required such as aerospace, racing sporting goods, pressure vessels, and other commercial products [15].

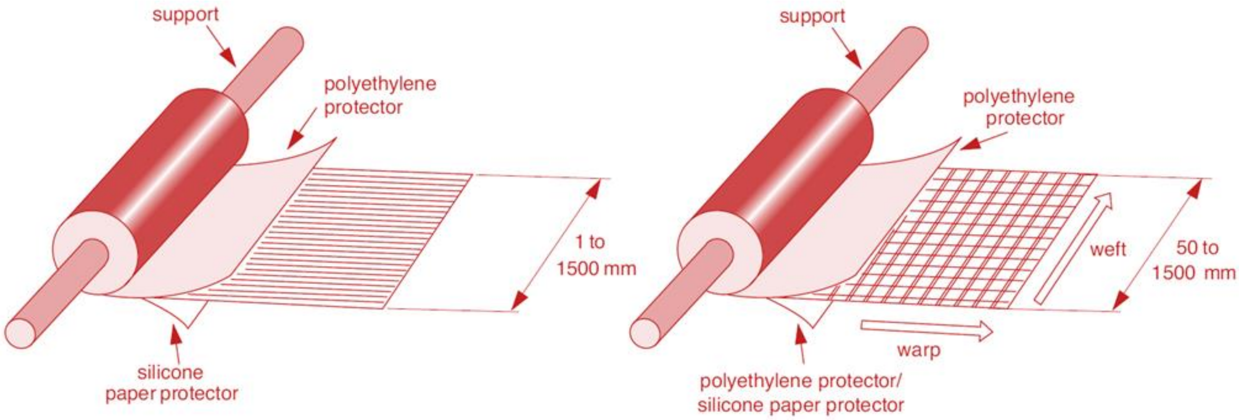


Figure 2.2 Prepregs in unidirectional and fabric form [1].

In the early 1980s prepregs were considered specialty materials, accounting for around 5% of an aircraft design and used only for non-critical secondary structures. Today, prepregs are the baseline for aircraft primary structures and constitute more than 50% of the airframe of the Airbus A350 XWB and Boeing 787. The growth in aerospace and other industries including wind energy, automotive, sports goods, and industrial machinery has followed. More recent applications benefiting from prepregs include subsea tubes for oil and gas exploitation and high-pressure vessels. This growth in the use of prepreg composites over metal has been driven by higher strength-to-weight performance, better fatigue strength, and the potential to offer greater freedom of design.[10]

2.3 Thin-ply technology – thin-ply prepregs

Among fiber composites manufactured with prepregs, thin ply composites made of plies of reduced thickness (from 10 to 150 µm) represent a promising technology for optimizing even further the performance of Fiber Reinforced Polymers (FRPs); they have gained both academic and industrial interest due to their unique combination of properties (FRPs) [16] [17] [18].

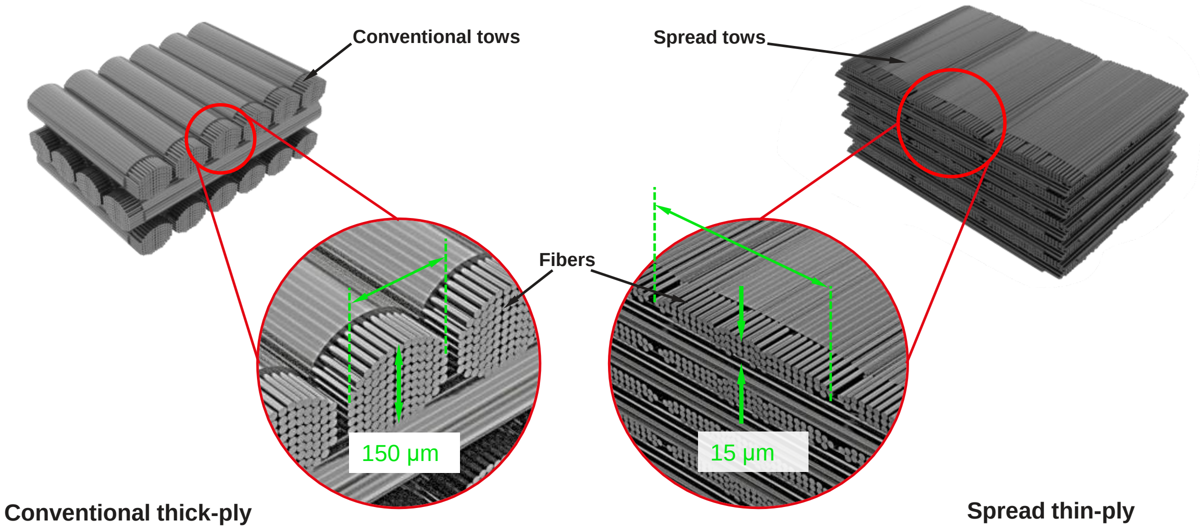


Figure 2.3 Schematic of convectional versus thin-ply prepregs.

They are now commercially available and have been successfully applied to manufacture light weight structures (such as the Solar Impulse plane), sailboats, but also aesthetic structures, as they allow either to form very thin quasi-isotropic structures (in the order of 100µm thickness) or to increase the material strength by limiting the damage zone, as will be presented in this chapter [19] [20].

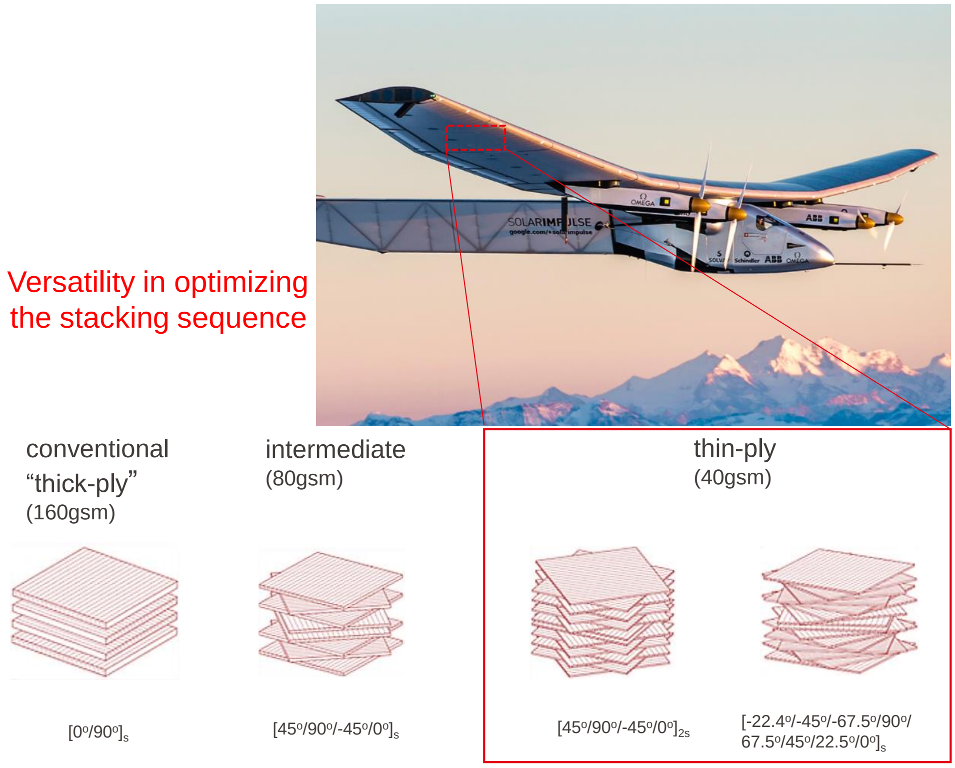


Figure 2.4 Thin-ply prepregs increase the design window for high-performance applications. adapted from:[21] [22].

2.4 Manufacturing of thin-ply prepregs

2.4.1 Fiber tow spreading and impregnation

Fiber tow spreading technology or thin-ply technology refers to the state-of-the-art composite preforming practices of spreading high-count bundles of technical fibers, typically consisting of more than 12.000 filaments (>12K) into a thinner, flatter, and wider unidirectional (UD) tape format. Remarkably, these processes can achieve a weight reduction of up to 500% per unit are of the initial “heavy tow” [23]. These thin UD tapes of reinforcement are then combined with a resin matrix system to form a thin-ply pre-impregnated material (thin-ply prepreg).

Although the concept of producing fiber composites with thinner layers has been explored since the late 1990s [24] [25] [26] [27], it was not until 2006 that significant advancements were made with the industrialization of tow spreading and impregnation processes. This industrialization enabled the mass production of homogenous thin-ply prepregs at a larger scale, resulting in a substantial reduction in production costs, making it a viable option for various applications of the composite industry. Moreover, it was during this period that research efforts began to specifically focus on the development and study of thin-ply materials [19].

2.4.2 Fiber spreading mechanisms

The industrial tow spreading processes and equipment are often protected by patents and considered proprietary. While specific details provided by composite manufacturers and suppliers are limited, there have been some insights provided in the academic bibliography or by analyzing production stages and spreading equipment through access to the patent’s bibliography. The work of Irfan et all [28] references various patents and studies and reveals some of the basic mechanisms that can be employed for the design and optimization of a fiber spreading apparatus. Additionally, the work of Gizik et al. [29] categorizes spreading methods in three general classes namely: active, passive, or a combination of both types.

Active methods involve using some form of energy to spread the fibers. Depending on the mechanism by which energy is offered to the filaments to flatten the tow, the active spreading methods can be distinguished as mechanical [30] [31] [32] [33] [34] [35] [36], electrostatic [37] [38] [39] [40] [41], pneumatic-vacuum [26] [42] [43] [44] [45] [46] [47] [48] [49], vibration [50] [51] [52] [53] and acoustic [54] [55].

Passive methods, on the other hand, rely solely on tension and constant movement to draw the fibers over spreading bars, convex, or other geometrical guiding elements to achieve a uniform spreading and reduction of the tow thickness. Frequently, combinations of more than one spreading method are applied to achieve the desired prepreg characteristics thus each method may allow different controllability over the process [29].

The experiments required for the development and production of all the types of (hybrid and non-hybrid) thin-ply prepregs were conducted with NTPT fiber spreading and impregnation methods. Although the technology of NTPT is proprietary, the three main production steps and the elements of NTPT's prepregger which consist of: the tow feed spools with an embedded tension control device, the spreading unit, the impregnation unit, and the take-up spool for the final product can be identified in Figure 2.5.

During the first stage fibers are passed through a pre-tensioning system with multiple rollers where large tows are dismantled into smaller sections. The second stage is the main spreading zone where energy is offered to the filaments in order to disassemble them into (ideally) individual filaments. At the last stage, fibers are guided to the impregnation unit and finally, the thin-ply prepreg is rolled on rotating drums and is available for use.

These production steps and all the modifications and new processes and equipment that allowed the production of hybrid prepregs will be analyzed in detail in Chapter 4, Chapter 5, and Chapter 6.

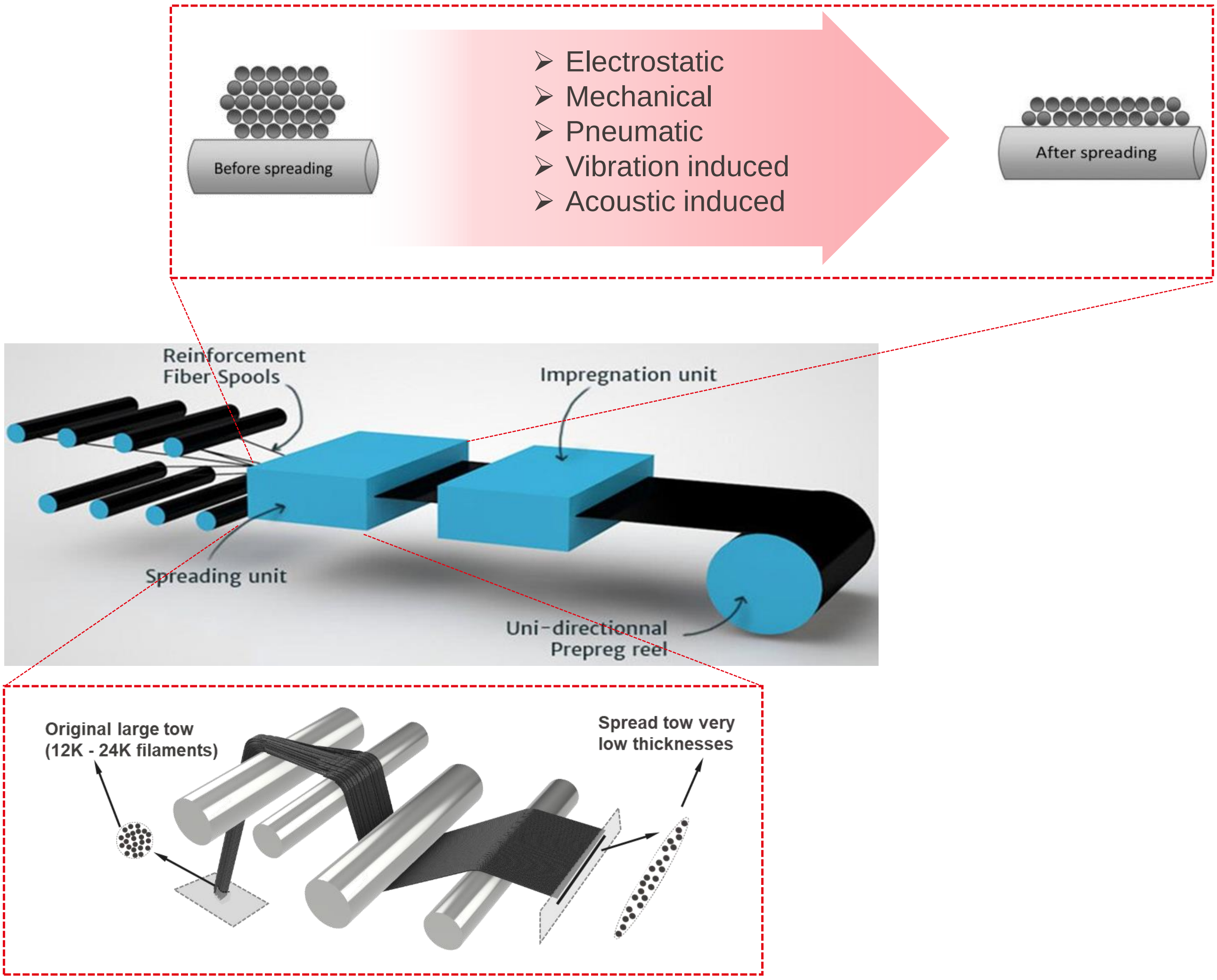


Figure 2.5 Schematic of NTPT thin-ply prepreg (fiber spreading and impregnation) production line Key fiber spreading mechanisms employed for the manufacturing of thin-ply prepregs.

2.5 Advantages and limitations of thin-ply materials

The first and most profound advantage of using thin-ply prepregs is the improved homogeneity of the microstructural profile compared to conventional composites. This can be attributed to state-of-the-art production methods specifically designed to align and distribute the fibers efficiently. By utilizing these production methods, common microstructural irregularities found in laminated produced with thick-ply systems, such as fiber misalignments, areas with imbalanced fiber/resin, and high void content resulting from fiber rearrangement and resin flow during the viscosity phases of the crying cycles, can potentially be eliminated. [20].

However, the major advantage of thin-ply prepregs is the exceptional improvement of mechanical properties that they demonstrate compared to standard composites, due to better use of the full potential of the fibers in tension. Several studies report that the use of thin plies in composite structures can enlarge the ultimate tensile strength, delay the onset of damage, expand the life cycle by improving fatigue resistance, and enhance impact resistance.

The first systematic study on the mechanical performance of thin-ply composite materials was conducted by Sihn et al. in 2007 [16]. In their research, quasi-isotropic thin-ply (0.04 mm) and thick-ply carbon epoxy composites (0.2mm) were subjected to unnotched tensile, static, and fatigue loading and compression after impact tests. Thin-ply laminates exhibited higher ultimate strength (+10%) in unnotched tensile test. Additionally, thick-ply specimens showed significant damage before ultimate failure while the thin-ply ones remained linear elastic until the fracture load. Improved fatigue behavior and post-fatigue resistance of thin-ply were also observed thanks to the reduction of damage developed close to the free edge of the specimens. After 50.000 fatigue cycles, the thin-ply composite did not show any significant strength reduction, while the thick-ply composite failed at 30% stress. Open-hole tensile test of quasi-isotropic specimens followed the same trend: Thin-ply laminates demonstrated limited damage development around the hole and prevented the propagation of large delamination cracks. Finally, thin-ply specimens showed less delamination after impacted compared to thick ones and better stability in compression after impact tests. Overall, because of the reduced free-edge effect and delayed delamination, thin-ply composites exhibited improved performance.

Similarly, Yokozeki et al. [56] investigated several strength properties as well as the damage resistance properties of thin-ply carbon fiber/toughened epoxy composite laminates for the applicability of these prepregs to aircraft structures. They performed a comparative study on CFRP laminates with the same in-plane stiffness, total thickness, and layup ratios (ratios of 0, ±45, and 90 plies) using standard prepregs (145g/m2) and thin-ply (75g/m2) prepregs is in order to determine the benefits of the use of thin-ply prepregs for composite structures. Specimens were subjected to static tension tests, tension–tension fatigue tests, and compressive tests including non-hole compression strength (NHC), open-hole compression strength (OHC), and compression after impact loading (CAI). It was shown that composite laminates manufactured from thin-ply prepregs demonstrated superior damage resistance properties compared to those from standard prepregs. Thus, it is expected that the design limit of composite aircraft structures increases by using thin-ply prepregs.

Yokozedi et al [57] also investigated the resistance of thin-ply composites subjected to out-of-plane loading. While multiple cracks and a large amount of delamination led to the failure of the thick-ply laminate, fiber fracture occurred in thin-ply specimens. This study investigated the damage characteristics of carbon fiber/toughened epoxy thin-ply laminates subjected to transverse loadings. Quasi-isotropic laminates were prepared using both standard prepregs and thin-ply prepregs to examine the effect of ply thickness on the damage accumulation processes and a clear difference between the two types of specimens was identified. Finally, the reason for the difference in damage process was investigated using finite element analyses, and it was clarified that the accumulated delamination position has a significant effect on the fiber fractures during the indentation.

Wisnom et al. [58] showed a ply thickness effect on the strength of unnotched QI specimens by comparing sublaminate scaling and ply level scaling. These two scaling strategies give drastically different strength values. Unidirectional specimens showed a decreasing tensile strength with increasing specimen size, with a reduction of 14% over a factor of 8 change in linear dimension Quasi-isotropic specimens increased in strength with size when the thickness was changed by repeating the sublaminate stacking sequence, with a 10% increase from a single to four stacked sublaminates. However, strength decreased when the thickness was increased by changing the ply block thickness, with a 62% reduction from 1 to 8 blocked plies. None of the laminates reached the equivalent strength of the unidirectional material, indicating that transverse cracking and edge delamination caused premature failure in all cases.

More recently Amacher et al. [20] [59] carried out a broad experimental campaign in order to study the influence of ply thickness on the ultimate strength and the onset of damage of composites. Specimens of various constituents and over a large range of ply thicknesses (t=0.030 to 0,300 mm) were tested and the improved properties of thin ply composites were confirmed at both laminal and component (bolted-joint bearing test) level. Furthermore, a linear increase of the stress at onset of damage with respect to ply thickness was reported. In Figure 2.6 it is demonstrated that when using ultra-thin plies (30g/m^2), damage mechanisms can be delayed until the ultimate failure strain of the laminate. In this case, the specimens fail in a brittle manner, while the thicker ply laminates show extensive matrix cracking and delamination.

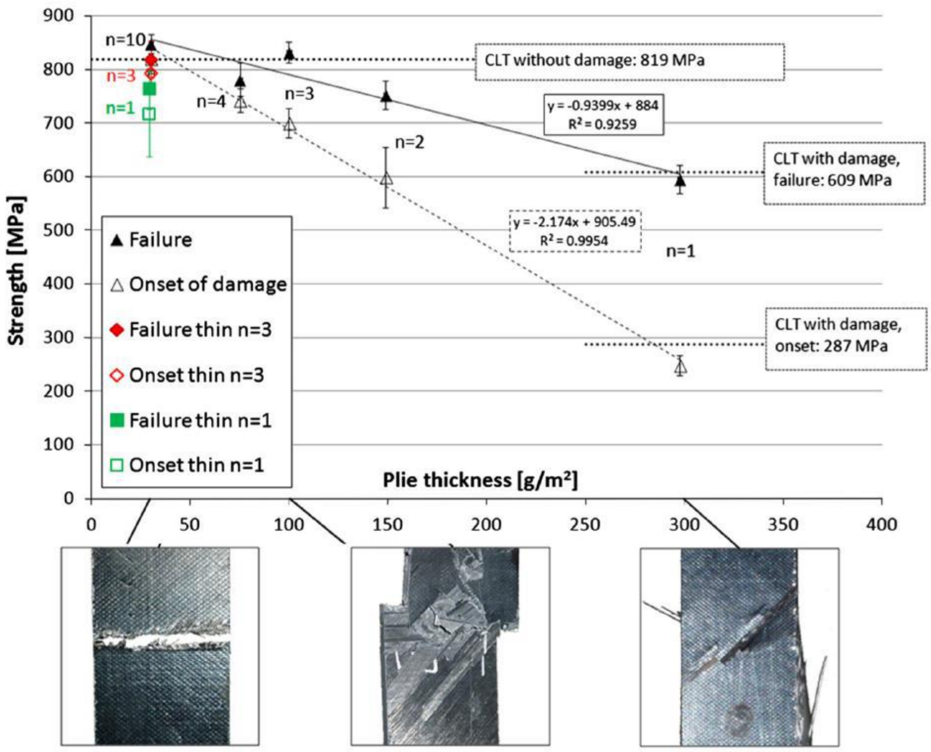


Figure 2.6 Onset of damage and ultimate tensile strength in unnotched QI specimen as a function of ply thickness [20].

However, the increase in stiffness and strength demonstrated in composite manufactured with plies of reduced thickness, resulting from delayed transverse cracking and delamination come at the expense of their limited toughness [60] [61] [62]. Therefore, any critical composite structures are often designed with large values of safety factors, preventing full exploitation of the potential advantages of these high-performance materials.

2.6 Fiber hybrid composites

The term “hybrid composite” is generally used to describe a composite material containing at least two types of reinforcement in its matrix. The purpose of hybridization is to construct a new composite material that will retain to the largest extent the advantages of its constituents and alleviate some disadvantages [63] [64]. Hybridization can also lead to synergetic effects or to properties that neither of the constituents possesses [7].

Research on hybrid composites started a few years after the invention of carbon fiber and was mainly driven by its high prices [65]: at that time hybridization was primarily an attempt to reduce the cost of composite structures by using cheaper glass fiber. Fiber hybridization is one of the simplest strategies that have been proposed over the years to introduce an adapted mechanical behavior to composites, for example, a more gradual failure mode, and reduce the major drawback of composite inherent brittleness [66].

It was first studied in 1972 by Hayashi [67] who reported that the failure strain of carbon fiber plies in carbon/glass hybrid composites was 40% higher than in the reference carbon laminates. Recently, Swolfs [66] reviewed the basic mechanisms responsible for hybrid effects and concluded that improvements for tensile failure strain on carbon glass/hybrids can range between 10-50%. Swolfs, [7] also highlighted that the magnitude of hybrid effects is highly affected by the degree of fiber dispersion and proposed the terms intrayarn, intralayer, and interlayer in order to categorize the three main hybrid configurations, as shown in Figure 2.7. In industry, these three configurations are also referred to as fiber-by-fiber, tow-by-tow, and layer-by-layer respectively [63] [7].

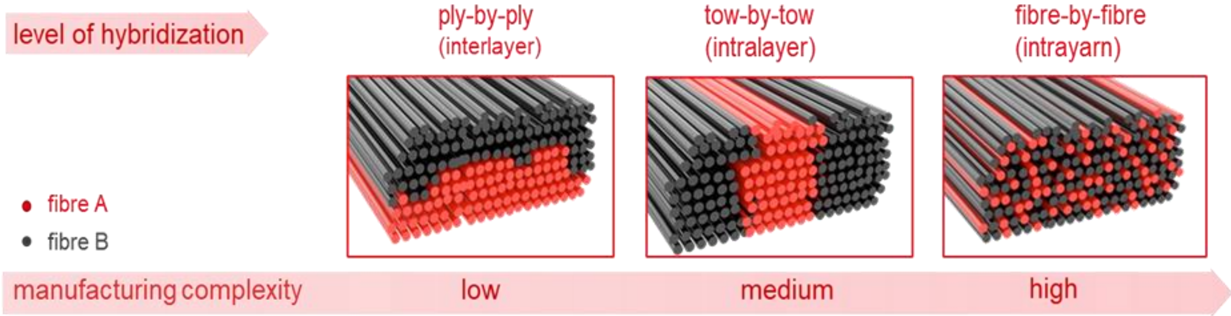


Figure 2.7 Three different fiber-hybrid configurations of unidirectional composites.

Ply-by-ply is the most straightforward hybrid configuration since it is relatively simple to design and can be achieved with already existing processing methods. For that reason, most of the early studies on hybrid composites were based on this architecture [68] [69]. Recently, ply-level fiber hybridization has been reported as an effective method to achieve pseudo-ductility [70]. Pseudo ductility is a metal-like nonlinear stress-strain response which can be introduced into composite structures by mixing low strain (LS) and high strain (HS) fibers at different levels.

Based on work showing the improved performance of single fiber (non-hybrid) thin-ply composites [16] [71] [72], Gzél and Wisnom [73] developed UD thin-ply glass/carbon hybrids in order to combine the advantages of hybrid and thin-ply effects in single laminate. They concluded that both the proportion and the absolute thickness of the constituent's layers have a strong influence on the ductile characteristics of the composite. The effect of geometric parameters on the pseudo-ductility of UD thin-ply hybrid laminates was also confirmed by the modeling work of Jalalvand [74] [75] [76] who developed the concept of damage mode maps, an efficient method that predicts the damage mode of UD ply level hybrids, gives accurate design information and illustrates once again the importance of thin plies for achieving optimum geometric parameters.

Guillaume Broggi, who also worked under the HyFiSyn framework, for his Ph.D. thesis on Multi-scale characterization and modeling of notched strength and translaminar fracture in hybrid thin-ply composites based on different carbon fiber grades [77], extensively studied the concept of damage mode maps developed by

Jalalvand et al. in order to identify the fiber-ratios and geometry characteristics of fiber hybrids that had the potential to improve the translaminar toughness of hybrids. These hybrid characteristics guided key thin-ply prepreg manufacturing decisions as one of the objectives of the present research work was to manufacture and provide thin-ply prepregs for further analysis and testing, to scientific partners and collaborators of the HyFiSyn project.

2.7 Model predictions for hybrids with increased fiber dispersion

Nowadays, although a considerable amount of work indicates that even ply-level hybrids, when carefully designed, can bring improvements in composite performance, simulation tools predict a great potential for tow-by-tow and fiber-by-fiber configurations [78]. Swolf et al. computed that the failure strain of 50/50 carbon/glass bundle-by-bundle hybrids increases up to 7% for architectures with small bundles (10 fibers/bundle) and up to 9% for randomly dispersed fibers, as shown in Figure 2.8 and Figure 2.9. The effect was significantly higher (16%) for single-fiber layer-by-layer architectures even though these hybrids seem less dispersed. These results are so far purely based on models but show that significant improvement should be obtained if the microstructure is carefully controlled.

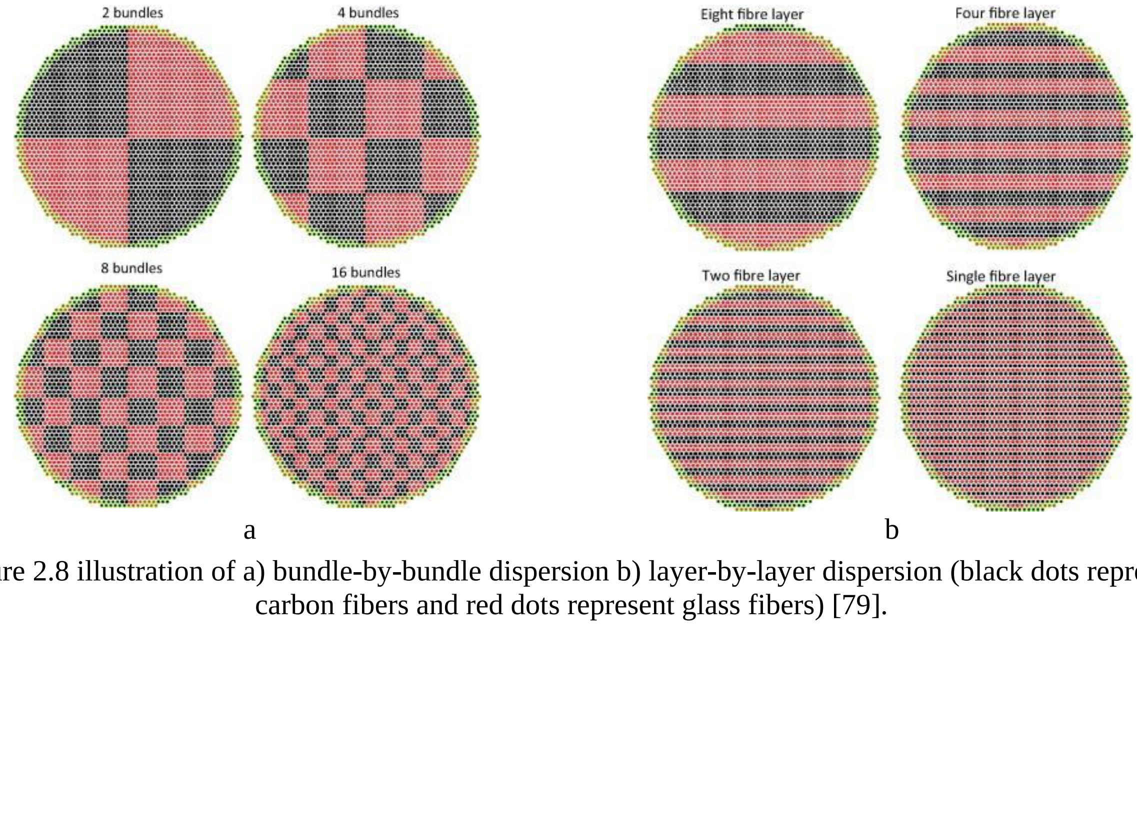
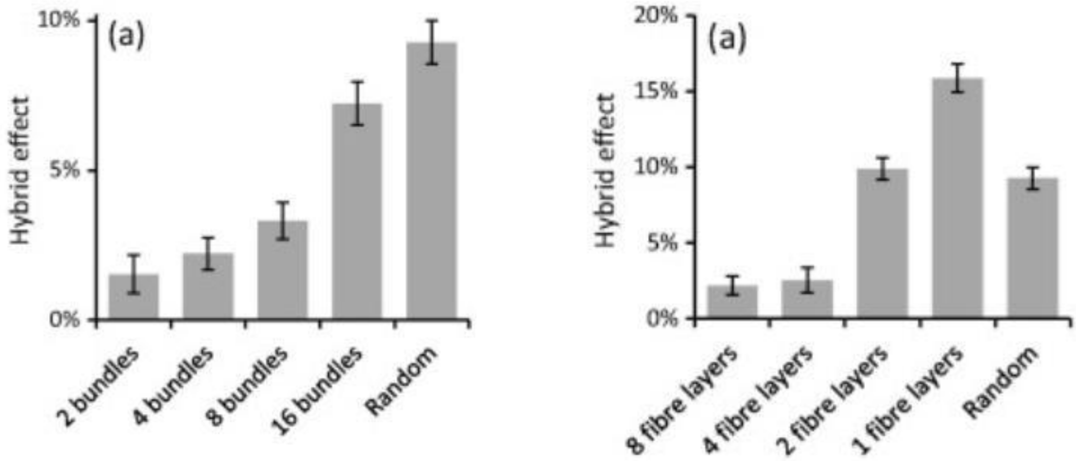


Figure 2.8 illustration of a) bundle-by-bundle dispersion b) layer-by-layer dispersion (black dots represent carbon fibers and red dots represent glass fibers) [79].



a b

Figure 2.9 Hybrid effect for a) bundle-by-bundle fiber dispersion b) layer-by-layer fiber dispersion [79].

2.8 Limitations of the current hybrid prepreg production methods

The industrial production of thin-ply prepregs that will enable the production of complex hybrid configurations still poses several manufacturing challenges since it is difficult and costly to combine different types of fibers, with different sizes, tensile behavior, and surface conditions.

Moreover, the influence of different processing methods on the microstructure of thin ply prepregs, and finally on the part microstructure, is still not fully understood, leading to composites where the microstructure is often an uncontrolled and not reproducible consequence of the processing route. Previous attempts at tow-by-tow hybridization have demonstrated the potential to arrange different fibers side by side in a hybrid pattern. However, the lack of control over other factors such as ply thickness and width of individual tows of different fibers has limited the ability to achieve consistent mechanical and microstructural performance. The SuCoHS (Sustainable & Cost-Efficient High-Performance Composite Structures Demanding Temperature and Fire Resistance) project, which investigated potential weight and cost savings in expanding the use of composite materials in areas of high thermal conditions (high temperature and fiber), was an example of one such attempt. North Thin Ply Technology managed to manufacture a tow-by-tow prepreg as a proof of concept. However, this prepreg was never tested for its mechanical and microstructural performance, and the focus was solely on achieving the hybrid pattern without paying attention to other parameters. In this thesis, we aim to build upon this work by achieving full control over fiber spreading and impregnation methods. This will allow us to produce new types of hybrid thin-ply prepregs with controlled fiber ratios and spreading ratios, enabling the development of high-performance materials with consistent mechanical and microstructural properties.

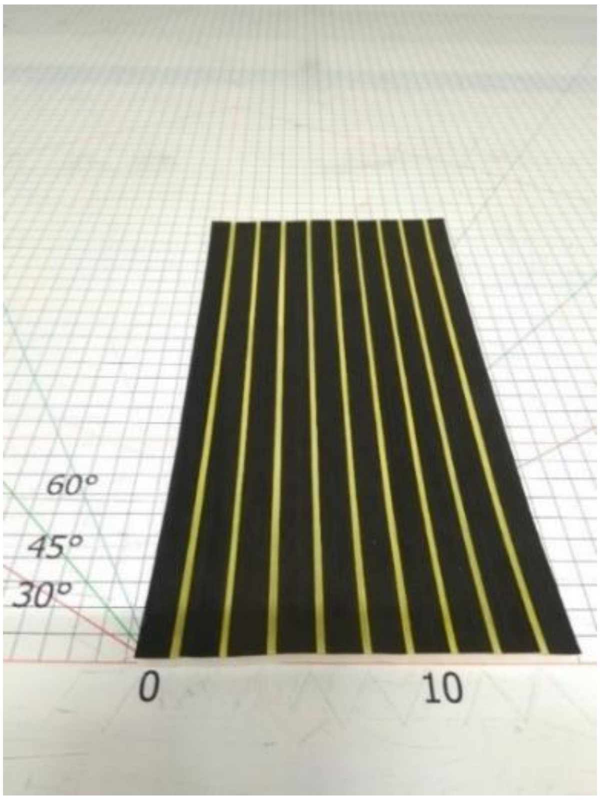


Figure 2.10 Proof of concept – early attempt for carbon/aramid fiber tow-level hybrid by North Thin Ply Technology source: NTPT.

The increased interest in the manufacturing of fiber hybrid configurations at both academic and industrial levels has been emphasized in recent research. As part of the EU ITN FiBreMoD (Fibre Break Models for Design novel composite microstructures and applications) project, which aimed to develop reliable design tools and predictive models for the mechanical properties of lightweight materials and fiber-reinforced composites, Ashok Rajpurohit performed research with Chomarat, a French company specializing in technical textiles, coatings, and composite materials, to develop and optimize a new generation of hybrid reinforcements focused on carbon and glass fibers at an industrial scale [80]. Due to the confidentiality of the industrial processes of Chomarat, the level of detail regarding the manufacturing methods that were applied is limited. However, during the FiBreMod conference, Rajpurohit presented some of the hybridization achievements resulting from this collaboration [81] Figure 2.11.

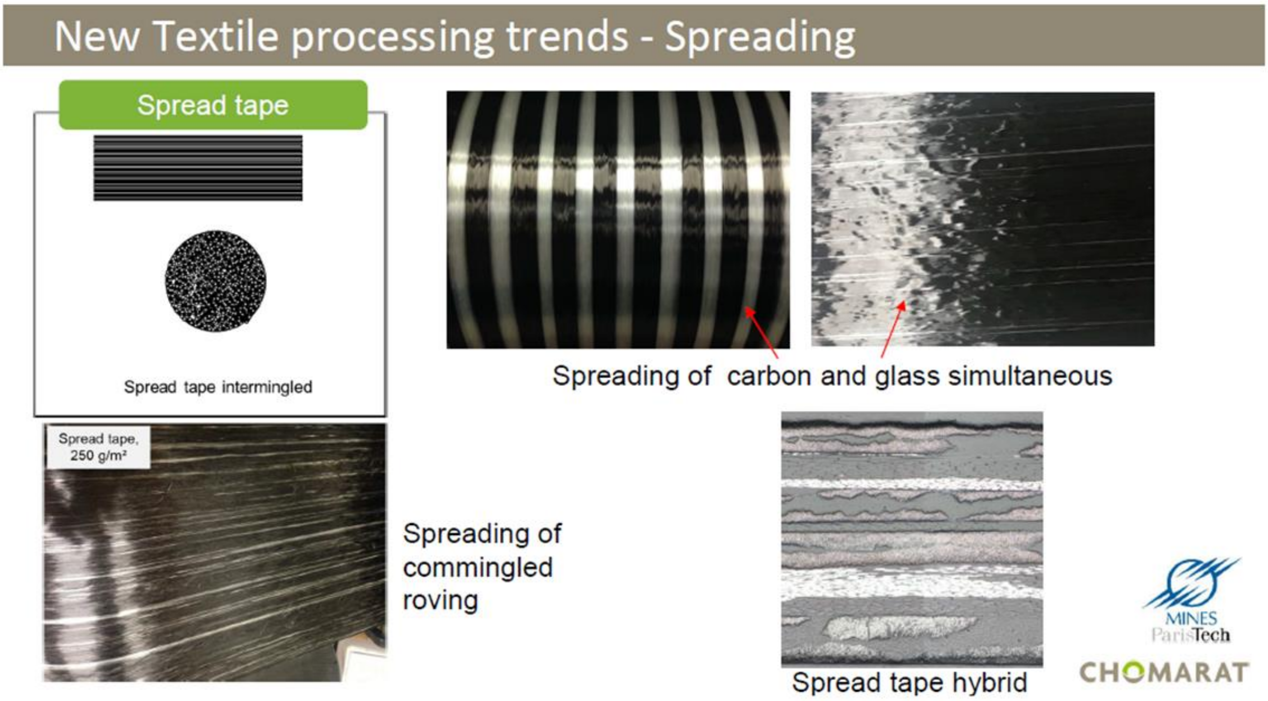


Figure 2.11 Thick-ply (250 gsm) carbon-glass prepregs for Chomarat [81].

From the results presented, it is evident that the process mainly relies on dry spreading, with no indication if the process can be adapted for the production of prepreg materials. This means that an additional step would be necessary for the manufacturing of the final part with the carbon glass hybrids produced from this work, and the benefits of prepreg technology cannot be fully utilized. Moreover, these hybridization methods produce thicker hybrid plies, thus they cannot be employed for studies an application that tries to capture thin-ply and hybrid effects simultaneously. Finally, there is a lack of information regarding the capability of the manufacturing methods to process two different grades of carbon simultaneously and not only carbon-glass. As a result, the hybridization methods' controllability is not fully understood, and the results may not be ideal.

These limitations highlight the current challenges and opportunities for further research and development in the field of hybrid composite configurations. By proposing a solution that addresses these limitations, such as a controllable hybridization process that utilizes prepreg technology and allows for the use of various grades of carbon fibers, the field can be advanced toward the development of lightweight materials and fiber-reinforced composites with improved mechanical properties. Additionally, Ashok Rajpurohit et Al. [82] investigated the potential carbon/glass hybrid composites for lightweight and structural applications and studied the effects of different inter- and intraply hybrid configurations on their mechanical performance.

All the different unidirectional (UD) reference and carbon-glass hybrids required for that study, see Figure 2.12 were developed with the so-called Malimo UD stitching technology. There is no mention of any fiber spreading mechanism and the tows of the different fibers were guided directly from the creel through a reed with specific dent spacing was used to control the required fiber areal weight and define the intraply pattern configuration by alternating the carbon and glass tows in the cases of tow-by-tow hybrid types.

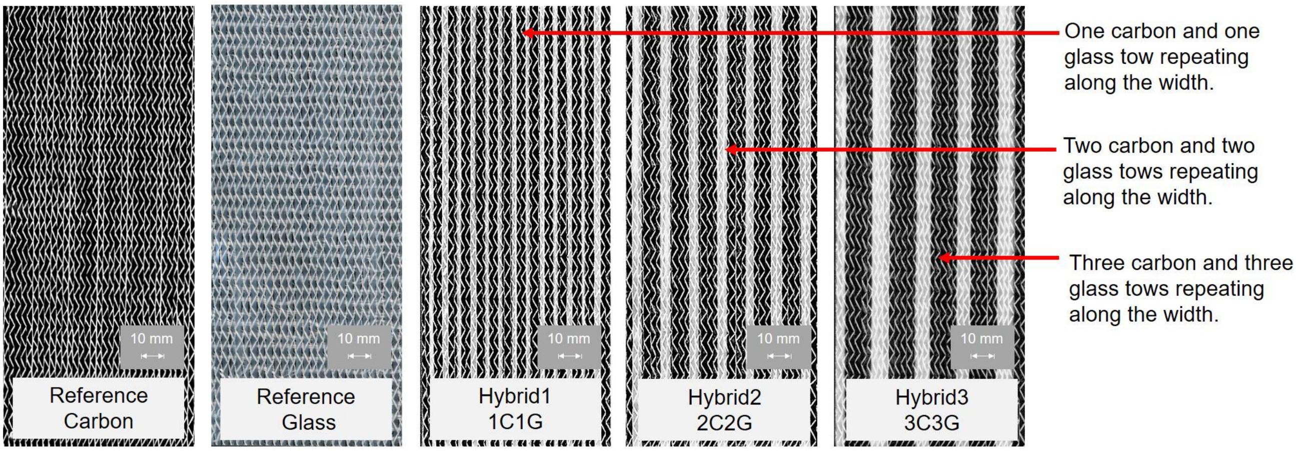


Figure 2.12 Photographs of developed fabric reinforcements reference and hybrid fabrics; and (b) magnified images of the face and back of a Hybrid1 fabric [82].

Furthermore, a stabilization fabric was introduced in the stitching zone, where this uniform stack of reinforcing fiber and the stabilization fabric were stitched together in a tricot stitching pattern using 170dtex Polyester threads Figure 2.13.

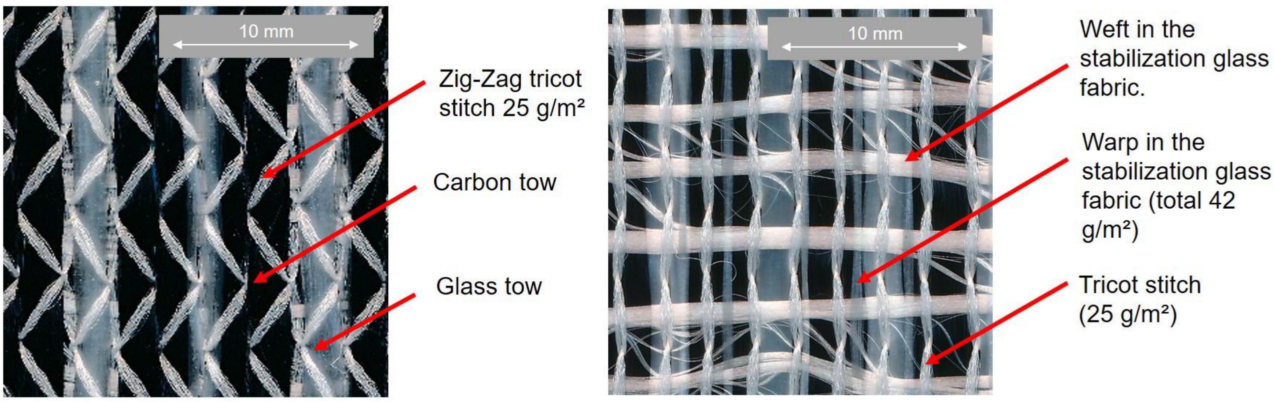


Figure 2.13 Photographs of developed fabric reinforcements: magnified images of the face and back of a Hybrid1 fabric[82].

The developed hybrid fabrics were used to create preforms and the researchers employed a low-pressure resin transfer molding (RTM) processing using epoxy for the manufacturing of composite materials.

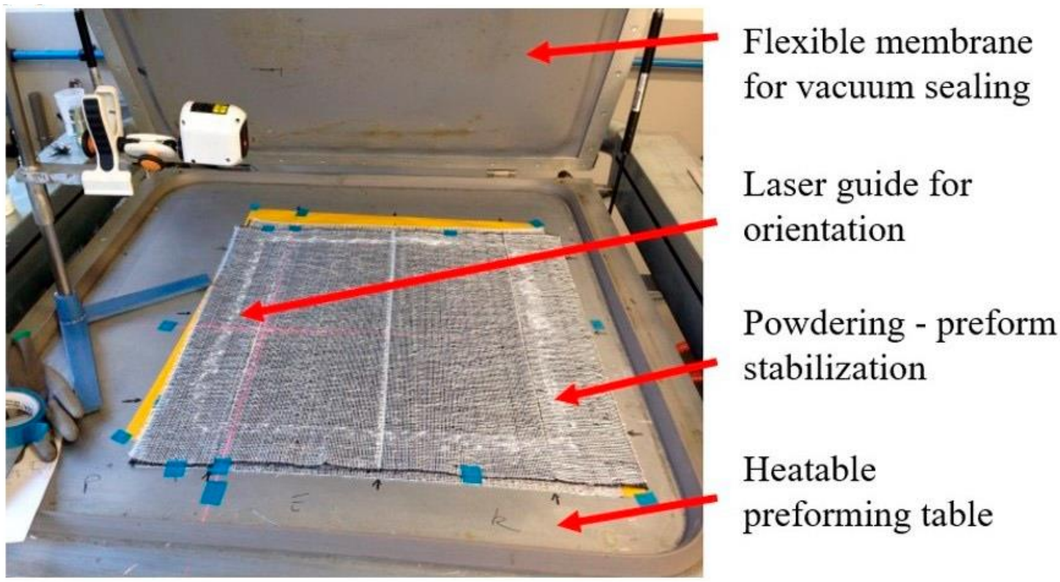
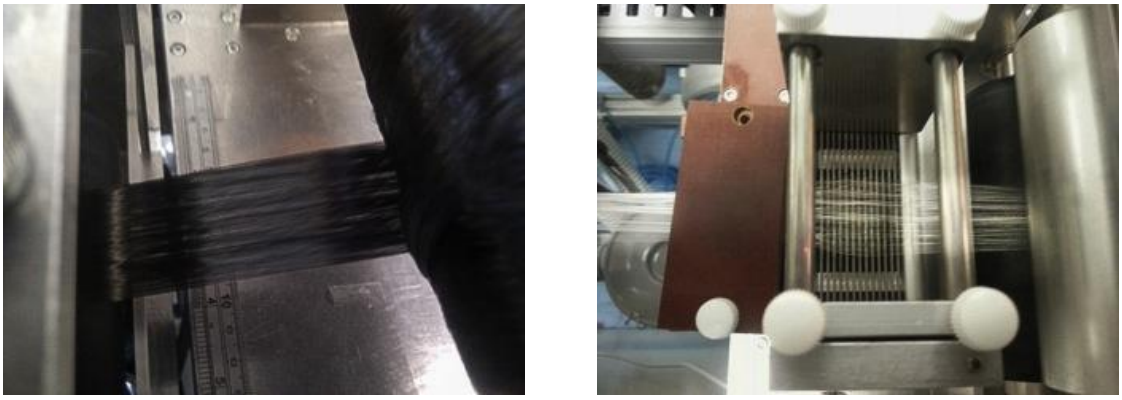


Figure 2.14 Preforming for hybrid composite manufacturing using RTM.

In conclusion, these manufacturing methods achieved some type of tow-by-tow fiber hybridization, but they relied on the addition of a stabilization fabric, which may have manufacturing and performance drawbacks. Additionally, the use of un-spread tows directly from the creels without any spreading mechanism and the stabilization of the produced hybrid configuration with a woven fabric with an areal weight of 42 gsm stabilization limits the achievable minimum thickness of the material. This thickness limitation means that the material may not meet the criteria to be classified as thin-ply material.

Moreover, the use of the RTM technique and stitching process also limits the potential benefits of using a prepreg process. Although Malimo UD stitching technology is used to develop unidirectional reference carbon and glass fabrics, there is no mention of fiber spreading, resulting in a thickness that does not meet the thin-ply composites category. Overall, the paper's methods can achieve either hybridization or thin-ply dimensions or prepreg technology, but not all together. The proposed innovation aims to create a material that can demonstrate hybrid thin-ply prepreg characteristics in a controlled manner.

Diao et.al 2014 [83] presented a novel method for producing a continuous intermingled hybrid composite material made of carbon fiber (CF) and glass fiber (GF). The authors used the principles of pneumatic/vacuum fiber spreading that lets air pass through the fiber tows in a low or no-tension state-based to increase the spacing between the fibers and thus the width of the entire tows compared as-received (high filament count -commercial tows). They employed a novel air-assisted fiber tow spreading technology to spread carbon fiber (CF) and glass fiber (GF) tows individually Figure 2.15 . These spread fiber tows were then intermingled using vacuum airflow Figure 2.15. This process allowed the fibers to be continuously intermingled to produce a continuous unidirectional hybrid (CF/GF) fiber tow containing 12K carbon and 1.63K glass fibers.



a b

Figure 2.15 Photos of spread (a) CF and (b) GF tows [83].

a b

Figure 2.16 Parts of the air-assisted where the co-mingling of the carbon fiber (CF) and glass fibers (GF) appearance of the combined GF/GF tows. (a) before entering (b) during commingling process inside the spreading unit [83].

Hybrid unidirectional hybrid composites laminates were manufactured with this new type of hybrid tow using resin film infusion for further testing. Microstructural analysis of cross-sectional areas of this new type of hybrids revealed the microstructural profile that can be seen in Figure 2.17. The authors reported a degree of dispersion of 42.45%, indicating only partial hybridization of the fibers,

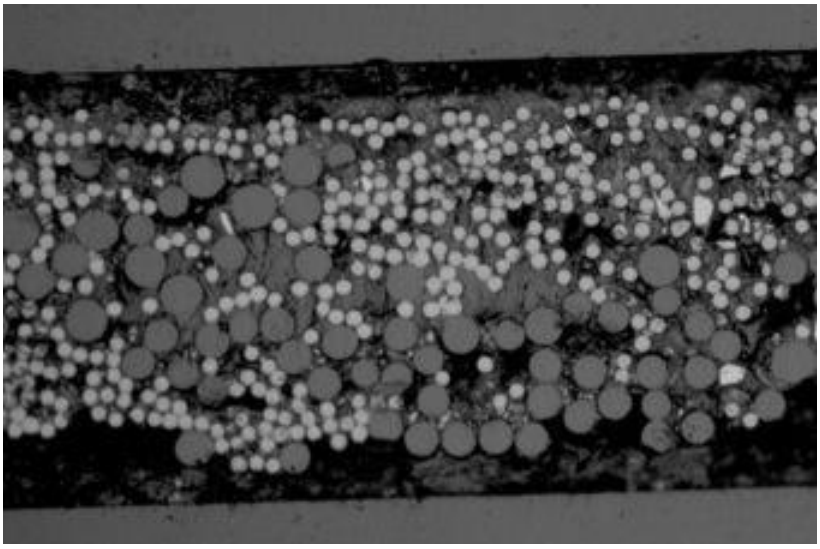


Figure 2.17 Typical micrograph of the hybrid CF/GF tow produced with the hybridization method of Diao et all [83].

Based on the information provided in the paper, it can be concluded that the air-assisted fiber tow spreading technology is a promising method for producing hybrid composite materials with improved mechanical properties compared to those made from carbon (CF) or glass fibers (GF) alone and a significant level of fiber comingling. However, it is important to note that the final properties of the composite material can be influenced by the resin impregnation and curing process so this type of material may not provide the same level of consistency as the use of hybrid prepregs. The reported fiber volume fraction in the produced composite material was relatively low. This may limit the practical applications of the material, particularly in high-stress applications where higher fiber volume fractions are needed.

Finally, R. Tavares [84] recently investigated the possibility to produce intra-ply hybrids of T800 and HR40 carbon fibers with an epoxy resin using a spread tow technology in a UD500 spreading machine from LIBA, at the Airbus Group Innovations in Munich, Germany. He explored the use of coated spreading bars, with added vibration and heat to add thermoplastic binders, and observed that the increase of spreading bars tends to increase the fiber tension, thus subsequent fiber damage. In addition, since the goal was to obtain intimate commingling of the two types of fibers, tow hybridization was performed after tow spreading using a second spreading unit, as shown Figure 2.18.

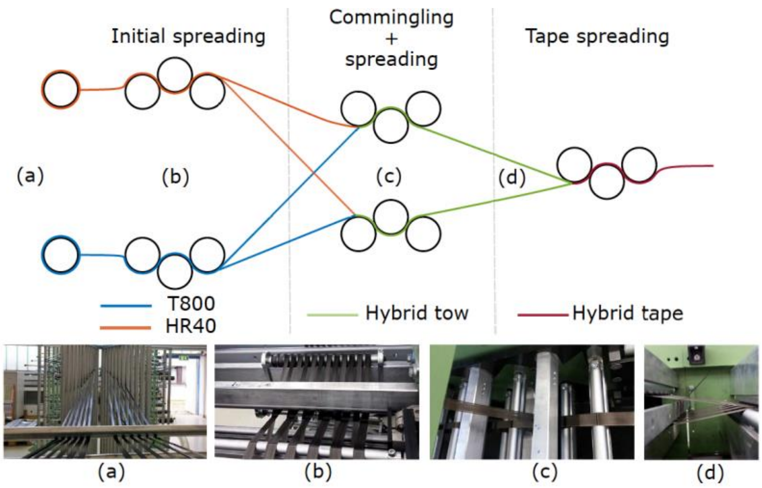


Figure 2.18 Schematic representation of the spreading process for hybrid materials and pictures of the different stages [84].

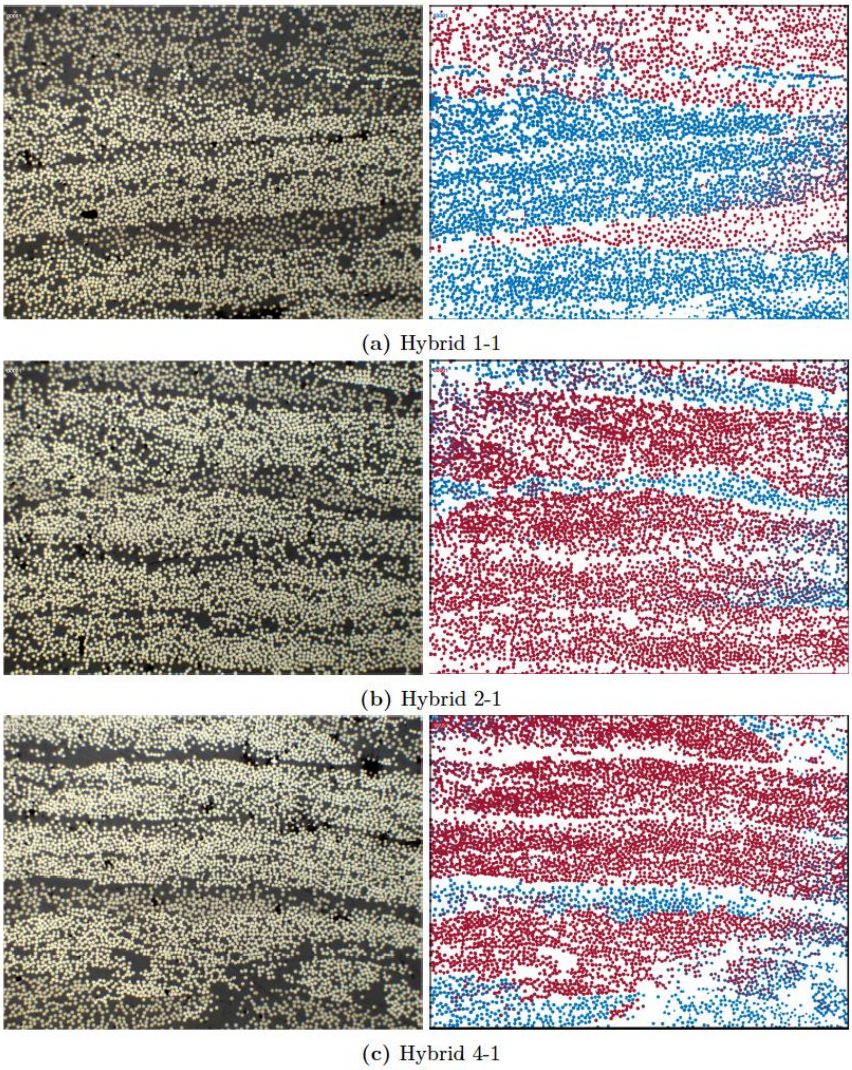


Figure 2.19 Microstructures and analysed microstructures of the hybrid materials in Tavares' work. HR40 fibers in blue and T800 in red [84].

The results were however not satisfactory, and the degree of spreading and mingling was poor, as shown Figure 2.19. Their main conclusion was that the initial spreading of the fibers should be better achieved, possibly with air spreading as performed by Diao, before any hybrid configuration can be envisaged.

Therefore, further research is needed to optimize the manufacturing process. Moreover, while the fiber tow spreading technology has the potential to be scaled up for industrial production, there are currently no records in the literature of this process being used on a large scale.

Despite these limitations, the authors suggest that the fiber tow spreading technology has significant implications for the design and production of hybrid composite materials with improved mechanical performance, which could be used in a variety of fields.

2.9 Microstructural characterization of composite materials

The microstructural analysis of fiber composite materials is an important aspect of research and development in the field of composite materials. This analysis provides insights into the structure, properties, and behavior of composite materials at the microscale, which is crucial for designing and optimizing the performance of hybrid materials [85] [86] [87] [88] [89].

Several techniques are available for the microstructural analysis of fiber composite materials, such as optical microscopy, scanning electron microscopy, and transmission electron microscopy. The choice of technique depends on the specific research objectives and the type of composite material being studied. Among them optical microscopy is an effective tool that is commonly utilized as the primary source of images in the investigation of the microstructural features and properties of fiber composite materials. This can be attributed to the combination of its ease of use and accessibility in both research laboratories and industrial settings and the numerous benefits it offers such as its non-destructive nature, high-resolution imaging capabilities, and real-time visualization. Additionally, the advancement of automatic image analysis techniques has significantly facilitated the characterization of microstructures in fiber composite materials. The continuous developments in this field, fueled by advancements in hardware, software, and imaging sources, have allowed for improved characterization of these materials, including the determination of fiber volume fractions, orientations, and the association of material behavior with fiber arrangements.

As the quality of the image is undeniably vital for the successful application of image analysis techniques sample preparation routines developed for polymeric-based samples [90] [91] have been tailored to overcome challenges arising due to the different nature of the constituent materials in the composite.

Composite materials often benefit from specific image enhancements, such as the removal of detected scratch marks and the separation of neighboring fibers that might have been identified as a single entity. Edge-recognition algorithms, such as those mentioned in references [92] [93] [94], play a crucial role in enhancing boundaries within the images. Enabling the detection of distinct features and morphological measurements applicable to composite materials.

However, the microstructural analysis of hybrid composite materials presents a unique challenge due to the presence of more the one type of fiber within a common matrix (at all stages of microstructural analysis, images, and processing description). Sample preparation of the cross-sectional areas of the composite can be particularly challenging, as the different fiber types may have similar physical characteristics, making it difficult to distinguish between them. This issue was addressed in this thesis by studying hybrids with two different grades of carbon embedded in the composite matrix.

A critical aspect in the development and study of fiber hybrid composites is the dispersion, which is a measure for how well the two different fiber types are mixed. It is defined as the reciprocal of the smallest repeat length [63] [69]. Swolfs et al. [66] provide a schematic illustration of the degree of dispersion in Figure 2.20. As shown in Figure 2.20 (a), a low degree of dispersion is characterized by two fiber types being in two distinct layers. To improve this, the number of layers can be increased or the layer thickness can be decreased, as shown in Figure 2.20 (b). Another approach to increase the dispersion is by hybridizing on the fiber bundle level, as show in Figure 2.20 (c). Finally, the best dispersion for this type of fiber hybrids in achieved when the two fiber types are completely randomly distributed as shown in Figure 2.20 (d)

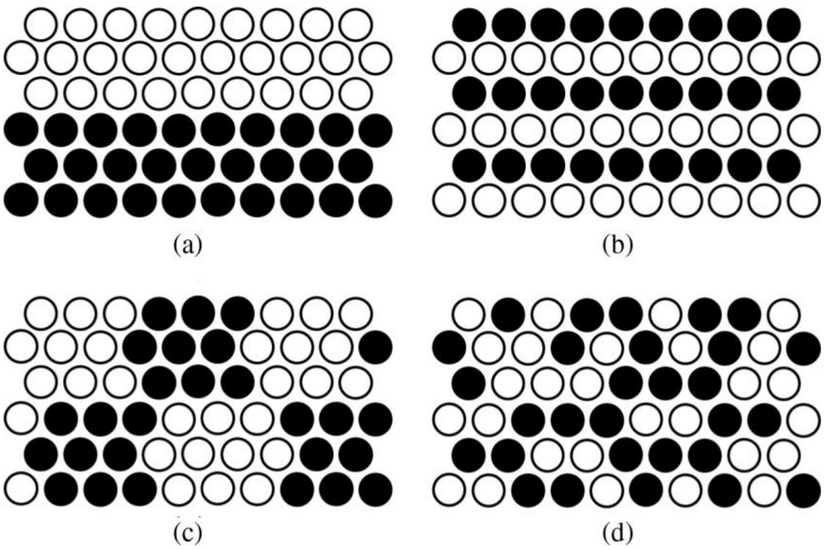


Figure 2.20 Illustration of the various degrees of dispersion (a) two layers, (b) alternating layers, (c) bundle-by-bundle dispersion, and (d) completely random dispersion [66].

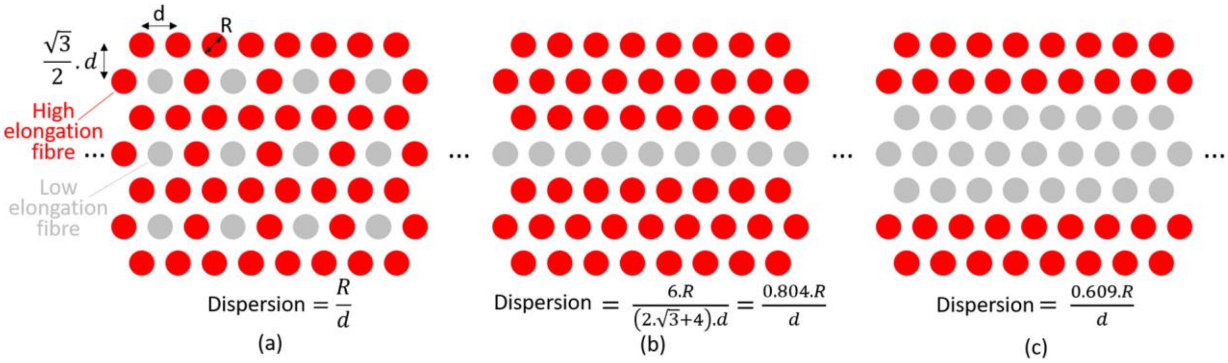


Figure 2.21 Schematic illustration of different fiber dispersions and definition for this term: (a) perfect isolation, (b) single fiber low-elongation layer and (c) a thicker low-elongation layer [79].

Diao et.al 2014 [83], proposed the term "degree of hybridization" and a methodology to evaluate the degree of hybridization in glass-carbon fiber hybrid composites. The degree of hybridization refers to the extent of intermingling between different fiber types within a composite material and served as a crucial parameter in quantifying the level of mixing and integration between the glass (GF) and carbon (CF) fibers in the hybrid composite manufactured with their novel air-assisted spreading method.

The degree of hybridization in the CF/GF tow was defined and characterized by comparing the fiber-to-fiber distribution obtained from a model of a composite containing randomly distributed two-fiber-types and the fiber-to-fiber distribution determined experimentally from micrographs of the fiber hybrid composite using a MATLAB image recognition program able to detect and measure the ration of glass to carbon fiber surface areas based on their image color intensity as shown in Figure 2.22

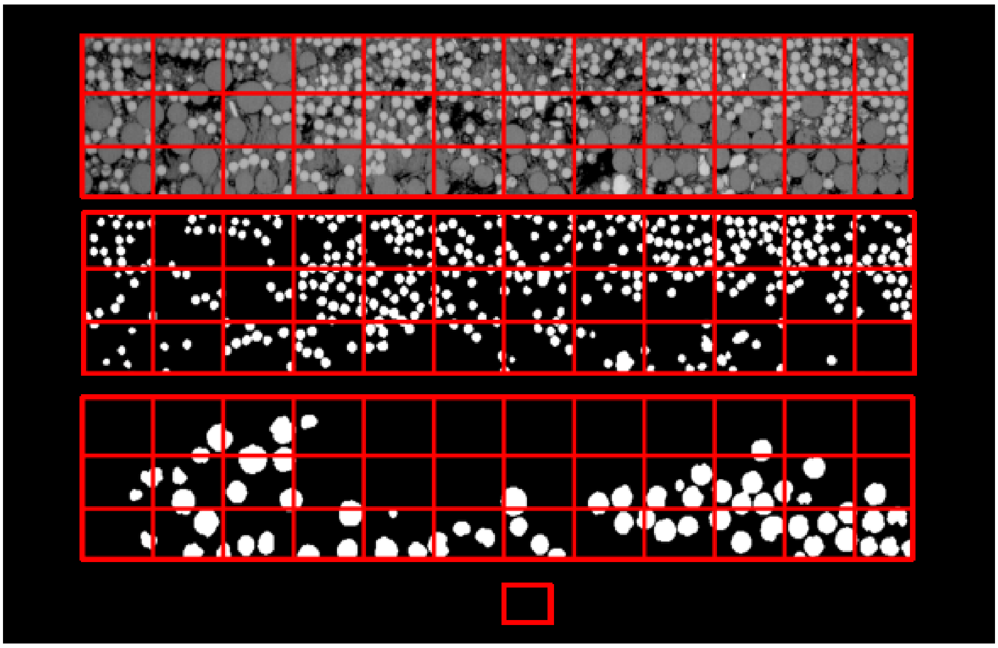


Figure 2.22 Schematic of the hybrid CF/GF tow image recognition program [83]

This work highlights that researchers can access the effectiveness of their fiber spreading technology in achieving the desired level of hybridization and their approach enables a quantitative assessment of the level of intermingling between the fibers that can contribute to the understanding of hybrid composite manufacturing techniques. However, it is important to note that although this work establishes a fiber-level composite hybridization process and a method to characterize the produced material, it does not specifically explore the correlation between the manufacturing processing parameters and the resulting microstructure of the hybrid composite. Building upon this foundation, my research aims to investigate the relationship between manufacturing processing parameters and the microstructure of hybrid composites. By exploring the correlation, we intend to provide further insight into optimizing the manufacturing processes and tailoring the microstructure to enhance the properties and performance of hybrid composites.

Finally, Katuim et al.[89] described a novel approach to identifying microstructural features in carbon fiber-reinforced polymer composites (CFRPs). The proposed method utilizes Voronoi tessellation, a mathematical technique, to evaluate the fiber volume content and clustering in cross-sectional micrographs of CFRP tapes. The approach considers the boundaries of the matrix and enables through-thickness analysis. The method is robust, suitable for automation, and has the potential to identify specific microstructural features and their relationship with processing behavior, thus eliminating the need for extensive manufacturing trials but there is no reference to its ability to identify more than one grade of carbon fiber within the same composite cross-section.

2.10 Role of the prepreg processing conditions on the fiber and composite properties

Very few studies directly relate the prepreg fabrication methods with the mechanical properties of the fiber and of the produced composites, and the present work aims to provide some means to fill this gap. Many studies, on the other hand, have been performed on the brittle behavior of single reinforcement fibers, with various types of treatments such as silane coating, and lead to evaluate the statistical nature of the fiber fracture using Weibull's statistics [95] . Performing a statistical analysis, for a given gauge length, of fiber fracture is now a very well-established method [96] for glass or carbon fibers. To predict the strength of composites, these probabilistic failure distributions are not sufficient as they do not capture the mechanism of stress distribution in composites, through the matrix and the fiber-matrix interactions.

Curtin [97] [98] [99] in the early 90’s, used the weakest link approach to propose a Global Load Sharing model GLS. This model was initially developed for ceramic matrix composites and extended to metal matrix composites. Recently, Rajan and Swolfs [100] [101] used this approach as a basis to predict the failure of hybrid composites, and R. Kumar, as part of the HyFiSyn project at DTU, also evaluated this model for carbon fibers [102]. The GLS model assumes that the matrix fails rapidly through cracking or yielding. The fibers hence carry the load, and the matrix role is to distribute the load to the other fibers upon failure, via fiber pullout, debonding and shear stresses through the matrix. The stress in the fiber is assumed to be recovered linearly over a critical length. In this zone along the broken fiber, the stress is hence lower, and no further break will appear. This area is called the exclusion or shielded zone. When enough fibers fail, the composite fails in a catastrophic mode due to damage accumulation. Kumar showed that this analytical model provides a practical method to link the fiber statistics to composite properties, however the failure strength predictions are in general about 30% higher than the real composite failure stress.

2.11 Conclusions of the state of the art

The state of the art in the field of industrial manufacturing of (thin-ply) pre-impregnated materials, fiber hybridization, and composite microstructural analysis techniques has been presented and several research gaps have been identified. While there has been increasing interest and significant progress in the field, previous attempts of fiber hybridization reveal that achieving a composite that combines thin-ply and hybrid characteristics with different levels of fiber dispersion in a controlled repeatable manner remains a challenge. In this context, this research work aims to address the current limitations and fill several research gaps by exploring the development of hybrid pre-impregnated composite materials. The increased controllability and the development of new processing methods and equipment will allow for the production of new types of hybrid thin-ply prepregs with controlled fiber ratios and spreading ratios, resulting in materials with improved quality and cost saving for the industry.

Firstly, the study aims to achieve hybridization at different levels by combining two different grades of carbon fiber within a single hybrid prepreg layer and benefit from the superior quality of prepregs, unlike previous attempts at hybridization in fabric, dry spreading, stitching, etc. (or added manufacturing steps). Secondly, the study aims to use two different grades of carbon fiber, rather than the commonly studied glass-carbon hybrids, to investigate the effects of fiber type on the microstructural and mechanical properties of the resulting material. Thirdly, several microstructural characterization techniques such as sample preparation, optical microscopy, and fiber identification algorithms will be explored to establish an effective way of revealing and studying the microstructural profile of carbon-carbon hybrid composites, which has been limited in previous studies. The microstructural analysis will also allow a correlation between the manufacturing processes and the resulting microstructure of the hybrid composite.

Chapter 3 Materials and methods

This chapter will first provide an overview of the key characteristics of the three different fibers and the resin system that were utilized for the manufacturing of all baseline (non-hybrid) and hybrid thin-ply prepregs throughout this study. Next, the prepreg processing methods and the manufacturing optimizations to produce high-quality multilayer thin-ply composite laminates, as well as the preparation of all testing specimens, are described in detail. Finally, the experimental techniques employed for the study of mechanical performance and the evaluation of the microstructural features of the thin-ply composites are presented.

3.1 Materials

3.1.1 Fibers

In this study, two distinct grades of carbon fibers were selected for the main production of thin-ply prepregs, namely the high-strain (HS) GRAFIL™ 34-700WD and the low-strain (LS) PYROFIL™ HR 40 12M. Both fibers were manufactured by Mitsubishi Chemical Carbon Fiber & Composites (MCCFC) USA and delivered in bobbins of 5000m in flat continuous tows designed for fiber spreading applications. Additionally, one type of Electric Glass (E-glass) Hybon 2026 from Nippon Electric Glass (NEG) Japan.

E-glass fibers were initially employed to study the effect of two prepreg processing stages, namely fiber pre-spreading and fiber thermal treatment, on single filaments and the overall quality of single-fiber (non-hybrid) thin-ply E-glass epoxy composite materials through a collaboration with HyFiSyn partners from Technical University of Denmark (DTU). All the experimental procedures regarding this part of the research will be discussed in detail in Chapter 4. E-glass was also utilized later in the project during the development of the fiber hybridization methods as a cheaper alternative to carbon. Its bright white color made easily distinguishable when mixed with carbon and allowed quick online adjustment that guided the process optimization before the stabilization of the hybridization processes and the transition to hybrid manufacturing with two different grades of carbon.

The E-glass was supplied on large 20kg bobbins of single-end round tows, designed for industrial use. Each E-glass tow consisted of approximately 6000 single filaments with a nominal diameter of 15 μm. The fiber sizing was adapted to epoxy resins, but the manufacturers provided no further information regarding its exact composition and content. The table below presents an overview of the key properties of all the fibers utilized in the current study.

Table 3-1 Characteristics of carbon fibers [103] [104] [105]

Fiber Reference	Fiber Type	Tensile Modulus [GPa]	Tensile Strength [MPa]	Elongation [%]	Price [€/Kg]
HR40	Low Strain LS	375	4410	1.18	69
34-700	High Strain HS	234	4830	2.06	15
E-Glass	High Strain HS	72	2186-2790	N/A ?	2-3

3.1.2 Resin System

The matrix was chosen to be the ThinPreg™ 415 (TP415), a proprietary rubber-toughened epoxy formulation of North Thin Ply Technology (NTPT), mainly designed for sporting goods applications. TP415 is recommended to be cured in an autoclave process to reach its optimal characteristics and performance. The properties of the resin obtained from the manufacturer's datasheet are gathered in Table 3-2. The experimental characterization of the properties of TP415 was carried out through macroscale experiments on specimens of neat resin at NTPT.

Table 3-2 Bulk matrix characteristics [106]

Resin Reference	Resin Type	Density	Tensile Properties			Flexural Properties			Tg
		ρ [g/cm^3]	E [MPa]	Strength [MPa]	σ [%]	E [MPa]	Strength [MPa]	σ [%]	On set [^{o}C]
TP415	Rubber toughened epoxy	1.208	3.01	75	4.89	3.3	138	75	147.1

The resin system has excellent compatibility with all the fibers of interest, which was confirmed with a preliminary testing campaign. The system's high elongation and great toughness can allow adequate shear stress transfer between the fibers and the resin. Of particular importance for the present study are the excellent processing characteristics of the resin system that is available in a thin-ply format and thus can promote thin-ply effects. The selected materials allow great design flexibility and enable the manufacturing of ultra-light unidirectional (UD) thin-ply prepregs with a fiber areal weight down to 15 g/m^2 (gsm). This offers unique versatility in real-life composite design. Finally, the stability of the resin and the long self-life increase the processing window. When stored sealed and out of direct sunlight prepreg materials impregnated with ThinPreg™ 415 (TP415) are expected to have the life indicated in Table 3-3. The extended out-of-freezer life of the resin was crucial for facilitating the advancement of hybrid prepregs mixing and processing techniques.

Table 3-3 Expected life for prepregs manufactured with ThinPreg™ 415 (TP415) [106]

Storage Temperature	Value	Unit
Shelf Life (-18°C)	24	Months
Out Life (+18-22°C)	8	Weeks
Tack Life (+18-22°C)	8	Weeks

3.2 Thin-ply prepregs materials

The selected materials were processed in-house at North Thin Ply Technology (NTPT) to produce all the prepregs that were required for the experimental characterization. During the process development phase of the study, the single fiber and hybrid thin-ply prepreg manufacturing was conducted in small batches on the laboratory-scale prepreg production line of NTPT in Renens, Switzerland. This enabled quick quality control of the preliminary hybridization attempts and online experimentation with different processing creating a loop between production and testing that promoted faster optimization of the hybridization techniques. When the optimal set of production parameters was defined for each hybrid prepreg configuration large batches of the required prepregs were manufactured at the production plant of NTPT in Zory Poland. The prepregs were delivered in unidirectional (width: 0.3 x length:50 m) rolls, sandwiched between two layers of backing paper that kept the fibers from sticking together, vacuum-sealed in polypropylene bags to be protected from environmental factors, and ready for further processing and composite manufacturing. When not in use they were stored under freezing temperatures (-18 ^{o}C) to reduce the reaction of the resin and catalyst that was initiated during the impregnation stage of the production and maximize their useable life. However, even at low temperatures some reactions still occurred, and in most cases, the prepreg material was considered unusable after 1-2 years.

3.2.1 Non-hybrid carbon thin-ply prepregs

Three types of single-fiber (non-hybrid) thin-ply prepregs with different ply thicknesses were manufactured with the HR40 and 34-700 carbon fibers. These thin-prepregs were first used as a baseline and for the manufacturing of ply-level hybrids. They were also employed as intermediate materials for the development of fiber-level hybrid configuration with the newly developed calendering process (see Chapter). The characteristics of the non-hybrid prepregs manufactured for this study are gathered in Table 3-4.

Table 3-4 Non-hybrid carbon thin-prepreg characteristics.

Prepreg type	Fiber Type	FAW [g/m^2]	Resin Type	Resin Ratio [%]	Sample Reference
non-hybrid	HR40	15	ThinPreg^{TM} 415	37	HR40 30 gsm
non-hybrid	34-700	30	ThinPreg^{TM} 415	37	34-700 30 gsm
non-hybrid	34-700	60	ThinPreg^{TM} 415	37	34-700 60 gsm

3.2.2 Non-hybrid glass thin-ply prepregs

Two types of single-fiber (non-hybrid) thin-ply E-glass prepregs were manufactured on the laboratory scale (R&D) prepreger of NTPT to study the effects of fiber spreading and impregnation production parameters on the mechanical performance of thin-ply hybrids. The existing prepreg production line was modified with the addition of custom-made parts namely the fiber pre-spreading stage and fiber thermal treatment stage and the prepregs were manufactured following several combinations of these stages. The exact processing routes and the working principles of the manufacturing method will be presented in detail in Chapter 4. Table 3-5 summarizes the characteristics of these E-glass thin prepregs.

Table 3-5 Non-hybrid E-glass thin-prepreg characteristics.

Prepreg type	Fiber type	Fiber processing production stage	FAW [g/m2]	Resin Type	Resin Ratio [%]	Sample Reference
non-hybrid	E-glass	Pre-spreading	50	TP415	38	E-glass PS 50 gsm
non-hybrid	E-glass	Pre-spreading & Thermal treatment	50	TP415	38	E-glass PST 50 gsm

3.2.3 Hybrid tow-by-tow carbon-carbon thin-ply prepregs

In total three types of tow-level (tow-by-tow) hybrid thin-ply prepregs were manufactured with HR40 and 34-700 carbon fibers; their production characteristics are summarized in Table 3-5. The new hybrid prepreg manufacturing processes that were developed for this study enabled the simultaneous spreading of dissimilar fibers in a single layer of a thin-ply prepreg tape are presented in Chapter 5. The production characteristics of all the tow-by-tow thin-ply carbon prepregs are gathered in Table 3-6.

Table 3-6 Hybrid tow-by-tow thin prepreg characteristics produced with simultaneous spreading and impregnation of dissimilar fibers.

Tow-by-tow hybrid	Tow 1	Tow 2	Resin	Ratio Fiber 1 / Total mass [%]	Tow 1: Tow 2	FAW [g/m^2]	Weigh resin ratio [%]
Type 1	34-700	HR40	TP415	27	1:2	90	37
Type 2	34-700	HR40	TP415	20	1:3	90	37
Type 3	34-700	HR40	TP415	27	1:2	45	37

3.2.4 Hybrid fiber-by-fiber carbon-carbon thin-ply prepregs

In order to investigate the challenging fiber-level hybridization four distinct types of fiber-by-fiber thin-ply prepregs were manufactured by mixing various ratios of HR40 and 34-700 carbon fibers utilizing a newly developed calendering process. The working principles of this novel prepreg processing method, in addition to the design details of the custom-made calendering apparatus employed in this study, will be thoroughly discussed in Chapter 6. Table 3-7 presents a summary of the characteristics and calendering processing parameters utilized in the manufacturing of the fiber-level hybrid prepregs.

Table 3-7 Hybrid fiber-by-fiber prepreg produced.

Calender Pressure	Low Strain Fiber (LS): HR40 [g/m^2]	High Strain Fiber (HS): 34-700 [g/m^2]	Hybrid FAW [g/m^2]	Sample Reference
Low	15	60	75	LP C 75
Low	15	30	45	LP C 45
High	15	60	75	HP C 75
High	15	30	45	HP C 45
Non-Calendered	15	60	75	NC 75
Non-Calendered	15	30	45	NC 45

3.3 Composite laminate manufacturing and specimen preparation

3.3.1 Prepreg lamination

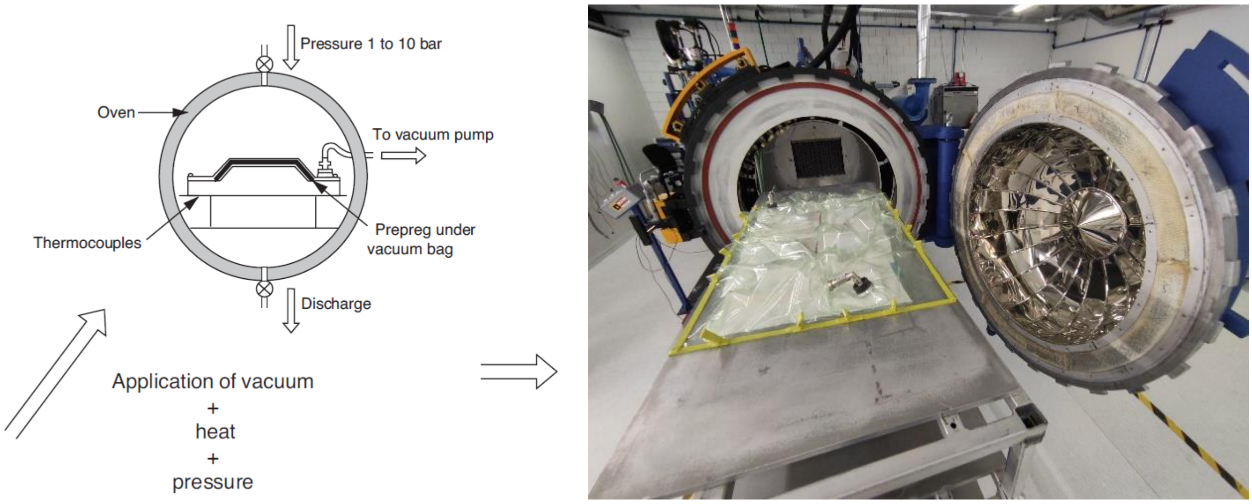
Before the initiation of the lamination process, the thin prepreg rolls were taken out of the freezer (-18 °C) and allowed to reach ambient temperature inside a vacuum-sealed protective polypropylene bag that was used for their storage, for 6-12h depending on the roll size. It was essential to avoid the appearance of any condensation on the surface of the prepreg rolls as any form of trapped humidity can significantly affect the quality of the laminates resulting in unacceptable porosity during curing and at some extreme cases even delamination of the plies. The non-automated hand lay-up lamination method was employed for the low to medium volume of production required for this study. These prepreg rolls could be delivered in an ATL (Automatic Tape Laying) ready-to-use state, simply by adjusting the position of the backing paper during manufacturing. However, hand-lay-up ensured full control of the lamination process and prevented imperfections and misalignments that could easily occur during the lamination of thin hybrid prepregs to produce complex hybrid architectures. The unidirectional (UD) tapes were stacked into large preforms of pre-stacked plies (4-16) of specific orientation sequence (UD: [0°], QI: [45°/90°/-45°/0°]) depending on the testing standard specifications. The preforms were then cut to the dimensions of the final plate. These preform ply-blocks were assembled on top of each other, and the number of them was adjusted to reach the targeted plate thickness.

3.3.2 Laminate debulking

During the prepreg lay-up process, it is common for unwanted air bubbles to become trapped between the prepreg layers which can later affect the laminate quality, and result in poor consolidation and increased void content. This unwanted air was removed by covering the laminate with a layer of breather fabric, and a vacuum bag and applying vacuum after the addition of each new ply layer or each ply-block. The vacuum was maintained for 5-15 minutes depending on the thickness of the prepreg layer and the total thickness of the laminate at room temperature. Longer consolidation and debulking cycles of at least 60 mins were carried out just before starting the curing process. This debulking procedure effectively facilitated good compaction between the thin prepreg plies and removed the trapped air which both eliminated the defects/void content resulting in homogeneous and high-quality laminates.

3.3.3 Autoclave curing process.

The thin-ply prepregs produced for this study can be cured following various manufacturing processes such as heated tools, ovens, and autoclaves depending on the quality requirements of the final application. The autoclave process was utilized as the most appropriate method for the manufacturing of excellent quality laminates containing high fiber volume and low void contents. Once the lamination and consolidation processes were completed, the laminates were properly vacuum bagged following the vacuum bag assembly shown in Figure 3.4 and placed inside the pressure chamber of the Akarmak (Turkey) autoclave of NTPT Figure 3.1. Inside the autoclave chamber the application of pressure and vacuum, the heat-up rates, and the curing temperatures were controlled throughout the process following the curing cycle recommended by NTPT presented in Figure 3.2.



a b

Figure 3.1 Autoclave set-up a) Schematic of a typical autoclave source: [10], b) NTPT autoclave.

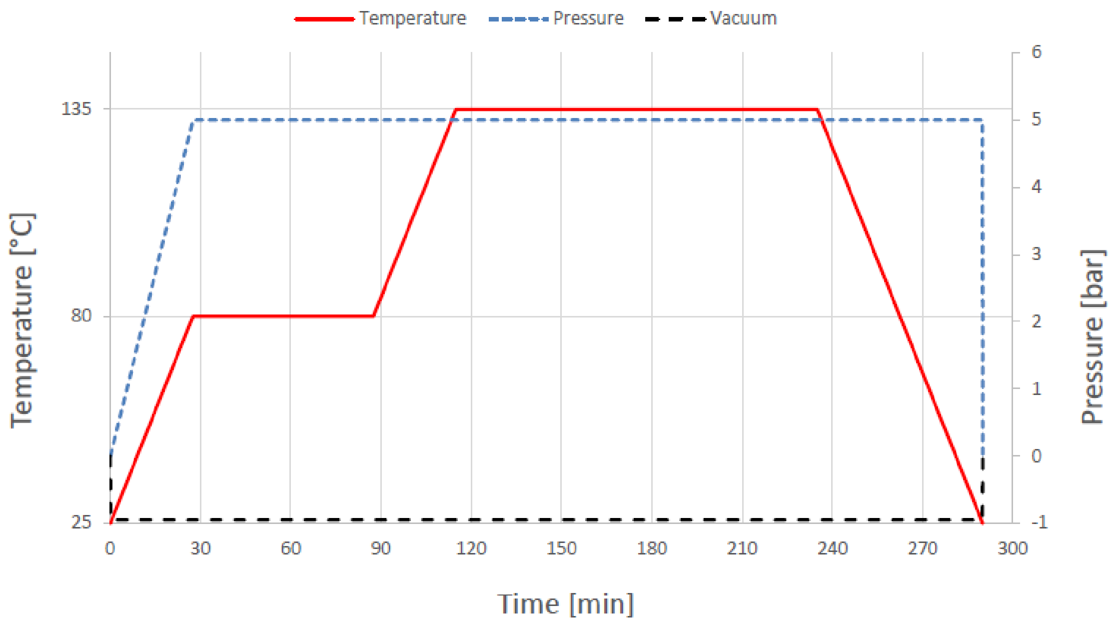


Figure 3.2 Curing cycle proposed by NTPT to fully cross-link the ThinPregTM 415 epoxy resin, achieve datasheet values and develop the best balance of properties for the composite laminate [106].

Long cure cycles were required because the large autoclave mass takes a long time to heat up and cool down. Sometimes slow heat-up rates are required to guarantee even temperature distribution on the tooling and composite components.

3.3.4 Optimization of the autoclave curing process.

In addition to temperature and pressure control of the curing cycle, the autoclave set-up and the use of specific consumables in the vacuum bag assembly were shown to have a significant impact on the quality of the cured laminates. Therefore, a series of preliminary tests were conducted to define the optimal autoclave set-up configuration that would produce consistent and repeatable quality results with regards to targeted fiber volume fraction (Vf: 55%), low void content (<1.5%), uniformity of thickness, minimal resin bleed, and smooth/bondable surface as presented in Figure 3.3. All the plates were cured by utilizing the setup illustrated in Figure 3.4 which combines the best characteristics of curing methods developed at NTPT and from previous work done at LPAC and the Laboratory of applied mechanics and reliability analysis (LMAF) of EPFL [107] [62]. These will be detailed below.

Figure 3.3 Curing multiple laminates with different curing set-ups to define the best-performing one.

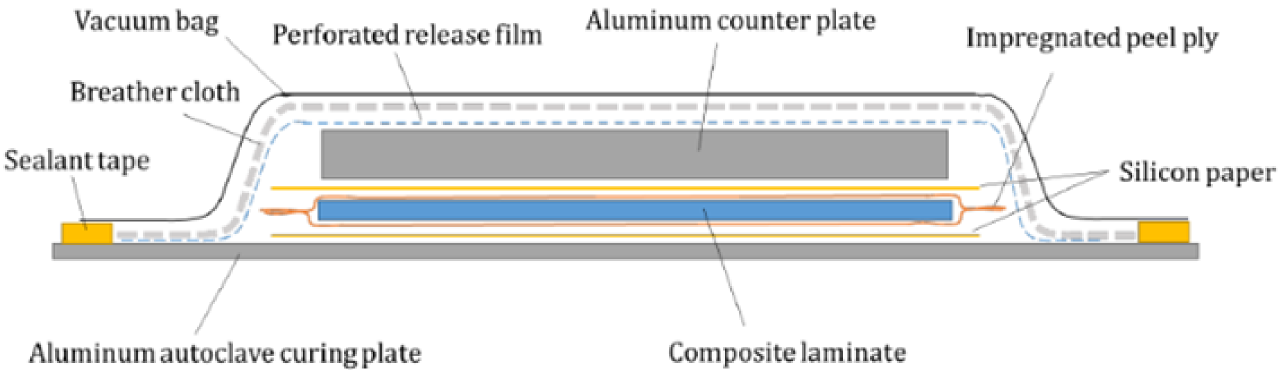


Figure 3.4 View of the different layers of curing consumables of the best-performing curing set-up and vacuum bag assembly.

3.3.4.1 Autoclave curing procedure and the role of the materials used.

The optimized curing configuration was employed to achieve high laminate quality. This included enveloping the laminates between two layers of peel ply impregnated with the resin used for the prepreg manufacturing (TP415, which minimized resin bleed by retaining as much resin as possible within the laminate during the curing cycle. Silicon papers were utilized as a release agent to facilitate the easy release of the cured laminate from the autoclave tool. An aluminum counter plate was employed to apply homogeneous pressure to the laminate and ensured uniformity of thickness and smooth finish on both surfaces and a perforated release film was utilized to allow for the release of air or any excess resin to flow and be absorbed by the breather cloth above. The breather fabric was utilized to facilitate the removal of air and void from the entire vacuum bag assembly while the vacuum bag and sealant tape provided the means to make an airtight bag on the surface of the autoclave tool. Two vacuum stems were inserted through the bag, with one directly connected to the vacuum pump and the other placed at the point furthest from the vacuum source to calibrate the vacuum gauges. Additionally, two control thermocouples were taped on the entire part which was positioned into the autoclave. Finally, the vacuum was applied to check for any possible leaks in the vacuum bag assembly or the entire autoclave vacuum system. Then the recommended curing cycle was followed. Upon completion of the curing cycle, the part was allowed to fully cool and then it was debagged and ready for further processing, such as specimens manufacturing as a post-curing was not necessary for this type of resin.

3.4 Microstructural analysis of hybrid thin-ply composite laminates

In this study, microstructural analysis was first used to verify the integrity of the produced thin-ply composite laminates and confirm that the targeted quality requirements were met before proceeding with further processing and testing. Hybrid prepreg tapes and laminates were evaluated in terms of homogeneity, impregnation quality, fiber volume fraction, voids content, degree of hybridization, and laminate quality homogeneity content of resin/fiber-rich areas.

Then, microstructural analysis on cross-sections of unidirectional laminates as show in Figure 3.5 was also utilized as the primary methodology for examining the level of fiber mixing that could be attained through various manufacturing processes and the effect of the production processing parameters on the overall quality of the thin-ply hybrid composite laminates. To achieve this goal, the processing steps of several different micrography specimen preparation procedures were combined and optimized to effectively address the quality variations arising from the different physical characteristics (fiber: type, diameter, color,) in a hybrid composite. Also, a new software for fiber recognition and extraction of statistical data from hybrid microstructures was developed. The following sections will provide a detailed description of the microstructural analysis techniques employed and how they enabled the study of the microstructural features of the hybrid composite materials and allowed correlations of the microstructural profile.

The overall objective part of the study was to investigate the correlation between the microstructural characteristics and mechanical properties of the materials and the manufacturing processing parameters used to produce them.

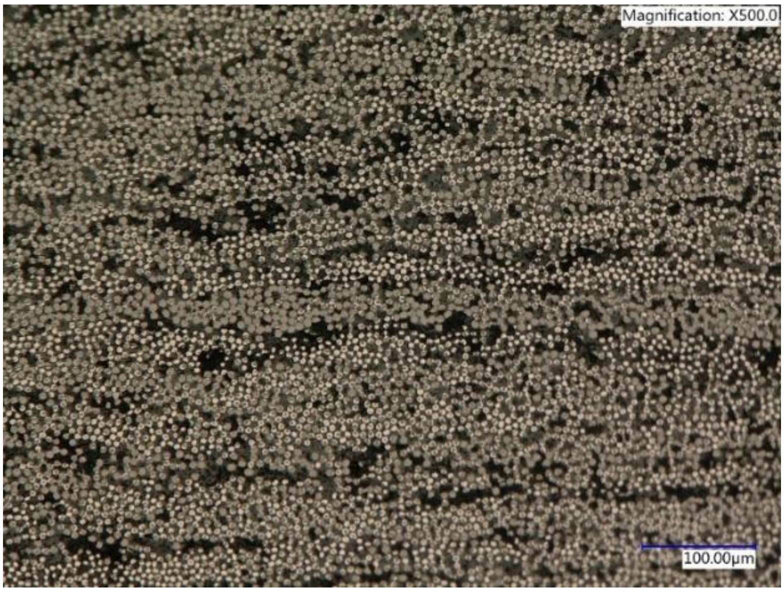


Figure 3.5 In the case of unidirectional (UD) fiber-reinforced composites, the microstructure corresponds to the fibers cross-section distribution. A typical microstructural profile of a laminate manufactured with 34-700 thin-ply prepreg.

3.4.1 Optimization of micrography sample preparation procedures – sample polishing

Cross-sections of the multilayer (hybrid and non-hybrid) autoclave-cured laminates were cut wet using a water-cooled rotary-blade saw to achieve undamaged edges. Starting from a clean and flat specimen surface is of great importance for the following steps of the specimen preparation process as it can reduce the grinding time and can help to better reveal the characteristics of the entire cross-section. The procedure steps are presented in Figure 3.6. The laminate cross-sections were then cast into resin blocks using a Methyl-methacrylate 605 LAM PLAN (France) fast-curing, cold molding resin. After the curing of the casting resin (30-1h), the blocks with the embedded micrography samples were ground with a SMARTLAM® 2.0 rotating polishing machine on LAMPLAN® silicon carbide abrasive paper discs. The grinding was performed under running water to lubricate the abrasive papers and wash away any debris particles to avoid surface scratches on the samples. The grain size of the discs started from P400 and after grinding cycles of 2-10 min, the process continued with finer papers of P400, P800, and down to P1200 respectively. During grinding a mechanical pressure arm was applying variable force (0-100N) on the center of the sample holder to avoid facets, the torque was constant, and the speed was set at 200 rpm. After grinding, the samples were finely polished to obtain a mirror-finish surface that allowed the different constituents of the hybrid composite laminates to be disguised. The polishing was done bi-directionally on a TOUCHLAM® polishing cloth that was lubricated using a Bio DIA-MANT® NEODIA® diamond abrasive suspension containing polycrystalline diamonds (diameters 1-3 μm). The polishing cycles for all the hybrid laminates were significantly longer (20-60 min) compared to non-hybrid (5 min) and were adjusted depending on the result's expectations. Finally, the samples were cleaned with soap and water and then rinsed with ethanol and dried with precision wipes.

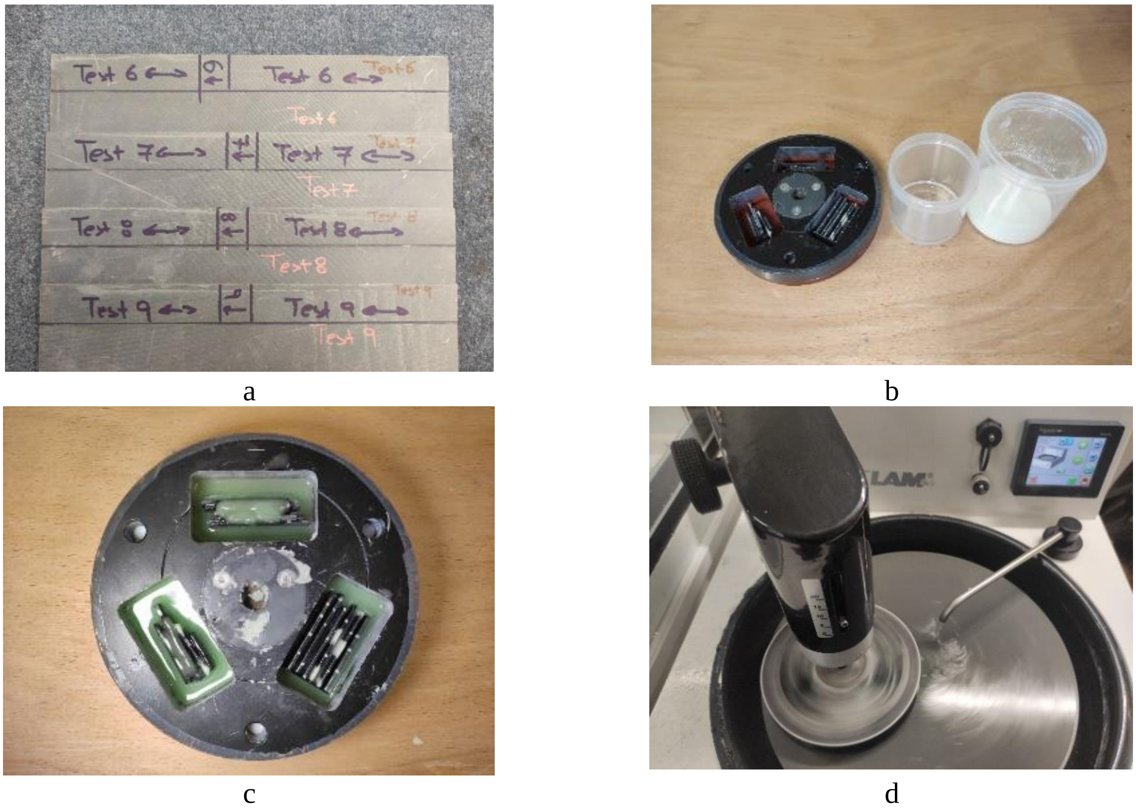


Figure 3.6 Preparation procedure of micrography specimens. a) marking and cutting cross-sections of the multilayer composite laminates b) mixing the two components of the casting resin c) placing the composite cross-sections into the mold of a polishing a polishing wheel and casting of the resin d) polishing on a rotary machine.

3.4.2 Optical microscopy

The micrography samples, consisting of polished laminate cross-sections, were examined with a KEYENCE VHX-600 (Japan) digital optical microscope. The specimens were observed at magnifications ranging from 100X to 1000X, enabling the detection and defects and microstructural characteristics at the level of individual fibers. The lighting conditions were carefully adjusted for each type of specimen using various light sources available on the microscope, including coaxial light, ring illumination, and mixed illumination. This allowed the achievement of an accurate white balance and the elimination of any unwanted glare, chromatic aberrations, and distractions. The aim was to capture crystal clear micrographs with high contrast between the matrix and the various reinforcement fibers, thus enabling the visualization of the microstructural features of the hybrids. The quality and resolution of the raw images produced at this stage of the study were critical, as they heavily influence the performance and accuracy of the fiber identification and image analysis techniques and software tool development for microstructural characterization of hybrid laminates

The integrated image analysis software the KEYENCE VHX-600 microscope provided some basic features for measurements on single fiber (non-hybrid) composites, that were used for the first quality screening.

However, its capability to perform measurements on hybrid composites was limited and the extracted data were unreliable.

To address these challenges, a new and versatile microstructural analysis approach was developed, specifically for thin-ply hybrid composites. This approach is based on a MATLAB tool for fiber identification capable of distinguishing multiple different types of fibers such as carbon or glass. Of great significance for this study though was its novel ability to recognize different grades of carbon fiber such as the HR40 and the 34-700 based either on geometrical or color (grayscale) characteristics. Additionally, the tool could provide input data for the artificial generation of representative microstructures and statistical analysis software and allow correlations of experimental observations done in the laboratory with ideal microstructures and statistical descriptors reported in the literature. This feature was utilized to quantify the level of fiber hybridization that could be achieved with each of the new hybrid prepreg manufacturing methods proposed in this study and also to evaluate the influence of the processing routes on the overall microstructural profile of hybrid thin-ply composite materials.

Finally, microstructural analysis techniques were employed, and a new image analysis tool was developed to capture the influence of specific production parameters on the overall microstructural profile of multilayer hybrid laminates (evaluation of the quality: ensured integrity of the plates, level of hybridization) and provide data for the development of the process models.

3.4.3 Fiber identification

The workflow for the identification of fiber position and type is shown in Figure 3.7. Matrix and fibers were segmented from the raw image in Fiji [108] via Minimum threshold. The following steps were conducted in Matlab. A morphological operation of opening was used to reduce thresholding artifacts prior to fiber identification via Circular Hough Transform. Analysis of the gradient magnitude on the location of each fiber was conducted on the raw image, leading to a bimodal distribution in which peaks correspond to each fiber type.

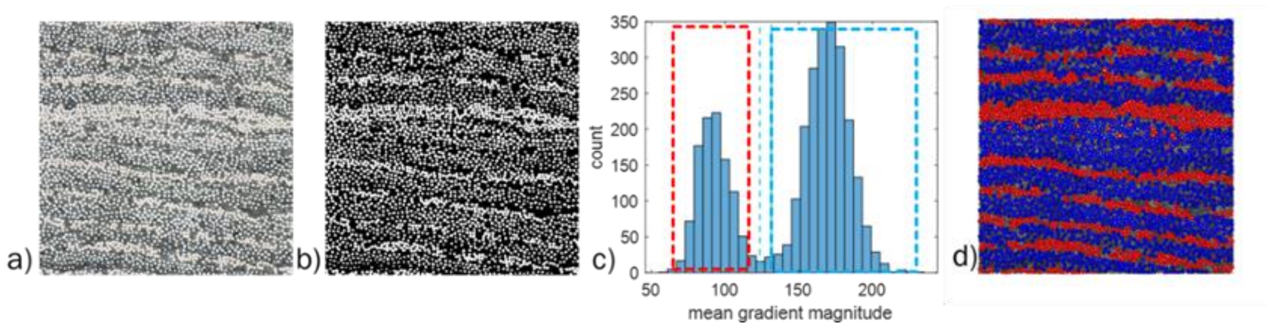


Figure 3.7 Workflow for fiber type identification: a) raw image b) image after thresholding and morphological operations c) histogram of the mean local gradient magnitude, showing a bimodal distribution where the two peaks can be associated to each fiber type (left: HR40 (low strain, LS); right: 34-700 (high strain, HS)) d) resulting fiber type assignment based on the bimodal distribution.

3.4.4 Hybrid parameters calculator

The metrics were based on the parameters of areal disorder [109], dispersion [110], and degree of hybridization [83]. To investigate the variability of the metrics for each material, the parameters were evaluated in three different regions of area 1600 µm by 1300 µm. Nominal fiber diameters were used for the calculations (7 µm for high strain (HS) fibers [111] and 6 µm for low-strain (LS) fibers).

3.4.4.1 Areal disorder

The areal disorder (AD) was determined using a Delaunay tessellation as defined by Bray et al. [109]:

$$AD = \left(\frac{A}{\sigma} + 1\right)^{-1} \quad (3.1)$$

where A is the mean area of the elements in the tessellation and σ is the corresponding standard deviation. In this work, AD was calculated for each sample on three different tessellations, one which considers globally all fiber centers and one considering LS fibers only and a third one considering HS fibers only.

3.4.4.2 Dispersion

In literature, the dispersion parameter was used to quantify the degree of fiber mixing by considering the proximity of two different fiber types $F_A$and F_B:

$$\text{Dispersion} = \text{mean}\left(\frac{R_{F_A}}{\sum_{i=1\dots6} d(F_{Aj} \rightarrow F_{Bi})/6}\right) \text{all j} \quad (3.2)$$

where R_{F_A} is the nominal radius of fiber type F_A, $d(F_{Aj} \rightarrow F_{Bi})$ is the distance between the j-th fiber of type F_A and the six closest neighboring fibers of type F_B. To understand the ability of the parameter to capture effects in an anisotropic fiber arrangement such as those encountered in this study, the parameter was calculated with reference to both low strain (LS) and high strain (HS) fibers, compared to only on low strain (LS) fibers as reported in the literature [110].

3.4.4.3 Degree of hybridization

The degree of hybridization compares the local fiber composition of a given microstructure to that of a microstructure with random fiber distribution [83]. A square moving window was used to determine the local areal ratio (AR) of the two fiber types over the region analyzed, as:

$$AR = \frac{N_{F_A} D_{F_A}^2}{(N_{F_A} D_{F_A}^2 + N_{F_B} D_{F_B}^2)} \quad (3.3)$$

where N_{F_A} and N_{F_B} are the number of fibers of the two types, and D_{F_A} and D_{F_B} the corresponding nominal diameters. The window size was chosen equal to six times the nominal diameter as suggested in the literature [83] and was determined as the weighted ratio of the nominal values of the fiber diameters in the mean fiber composition for the dataset. The degree of hybridization (H) was then defined as the ratio between the coefficient of variation of the areal ratio distribution of each sample and that of a random hybrid microstructure, expressed as a percentage. For this analysis, the random microstructure was chosen to have same fiber type ratio, fiber number, and positions as in each sample, but with a random fiber assignment.

3.5 Mechanical Testing

The investigation of the mechanical behavior of the hybrid thin-ply composites was conducted according to the ASTM D3039/D3039M [112] International standard on an axial-torsional MTS 809 tensile machine using a 100KN load cell. The strain was measured using HBM 1-LY41-6/120A resistive strain gauges glued on treated surfaces in the center of the sample's gauge section. The signal of the strain gauges was run through the strain-gauge amplifier integrated into the MTS FlexTest ® 40 digital controller used to drive and operate this mechanical testing machine. The tests were conducted in ambient conditions with a loading rate of 2 mm/min.

3.5.1 Specimen preparation for the tensile test (QI)

The specimens for the tensile test were obtained from quasi-isotropic panels with dimensions (570 x 300 mm). The stacking sequence was the quasi-isotropic (QI): [+45^o,0^o, -45^o,90^o]ns to stay aligned with previous work performed on QI laminates. Additionally, quasi-isotropic composite laminates are of great interest to North Thin Ply Technology (NTPT) as they are used throughout their product portfolio. Quasi-isotropic laminations are extensively used in the aerospace industry, especially in wings and fuselage panels, where their improved mechanical properties can lead to weight savings, increased structural efficiency, and performance, but also in automotive, marine, wind energy and sports applications which are all target markets for NTPT. The laminates were balanced and symmetric with a double 90^o ply lying on the sample's symmetry plane and they were manufactured from 2 mirroring quasi-isotropic preforms. The number of plies (n) was adjusted between samples manufactured with prepregs of different ply thicknesses to ensure a consistent specimen thickness of 2.5-3 mm across all the samples following the testing standard. This test type requires tabs for the gripping of the testing coupons. Tabs of 2 mm thickness from cross-ply CP: [0^o, 90^o]ns laminate configurations produced with unidirectional single fiber 34-700 60 gsm thin-prepreg in order not to waste the hybrid prepregs. The tab material was applied at 45° to the force direction to provide a soft interface. The tabs were prepared and bonded with a high-elongation (tough) epoxy adhesive and cured under vacuum in an oven on the material under rest [113] to form a tabbed laminate as illustrated in Figure 3.8. A uniform bond line of minimum thickness is desirable to reduce undesirable stresses in the assembly. To achieve clear and undamaged specimen edges a rotary saw with a water-cooled diamond blade was used for all the specimen cuttings and a custom-made tab alignment tool as illustrated in Figure 3.9 was employed for the tab bonding. Figure 3.10 presents the final geometry of the samples, and Figure 3.11 an illustration of the final set-up during tensile testing.

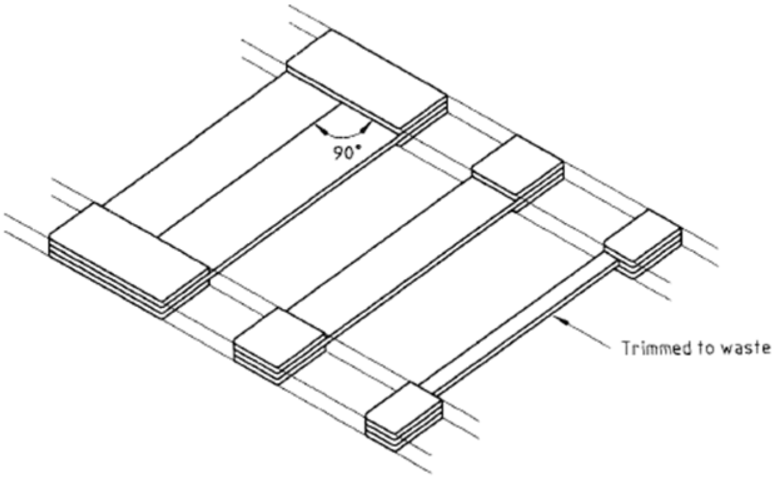


Figure 3.8 Schematic of a tabbed laminate for tensile specimen preparation [113].

Figure 3.9 Custom-made tab alignment device.

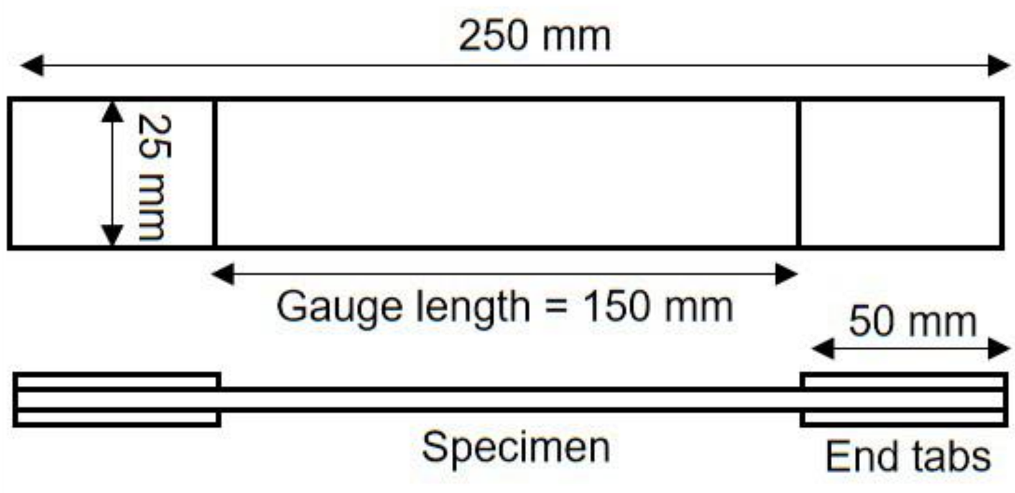
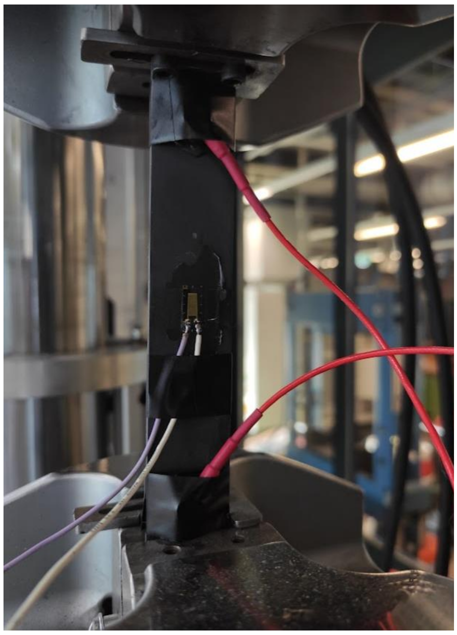


Figure 3.10 Tensile specimen dimensions (250 × 25 × 2.5 mm) according to ASTM D3039/D3039M [112].

a



b

Figure 3.11 a) Tensile testing set up b) Placement and alignment of the testing specimens in MTS 647 Hydraulic Wedge Grips of the mechanical testing machine.

3.5.2 Strain monitoring

The UNT tests were performed on an MTS 809 hydraulic tensile machine, under monotonic displacement, at a loading rate of 2 mm · min^{-1}. The strain was monitored with HBM 1-LY-41-6/120 strain gauges glued at the center of the sample with a cyanoacrylate adhesive (SCS Glue X2).

Chapter 4 Non-hybrid thin-ply prepreg manufacturing

This chapter provides a comprehensive overview of the thin-ply prepreg manufacturing methods, to aid pre-spreading of the fiber tows, and their effect on the final quality of the thin-ply prepreg material. Modifications and custom-made pieces of equipment that were designed, built, and implemented into the existing thin-pre-preg production line at North Thin Ply Technology (NTPT) will be presented. A case study on glass fiber thin prepreg will be presented, evaluating the influence of the spreading technique on both the single fiber mechanical performance, and the resulting composite performance.

4.1 Materials and equipment for thin-ply prepreg production

A variety of single fiber (non-hybrid) unidirectional (UD) thin-ply prepreg rolls with different characteristics were produced with the method that will be presented in this chapter. These non-hybrid prepregs were first used as baseline materials or they were manually mixed at a ply level to create simple ply-ply hybrid configurations. More importantly, they served as intermediate materials for the development of methods to achieve more complex hybrid architectures, such as tow-by-tow and fiber-by-fiber.

While the exact configuration of the production line may vary among different manufacturers, a typical thin-ply production line can be broadly divided into three main stages, namely: fiber feeding, fiber spreading, and fiber impregnation as illustrated in Figure 4.1. In addition to these primary stages, several other secondary stages may also be present to provide an online evaluation of the quality of the product and/or facilitate the handling of the final product and ensure a smooth and uninterrupted operation of the production line. The following section will discuss step-by-step the role of each production stage to provide a thorough understanding of the current industrial prepreg manufacturing methods. This is crucial to highlight the limitations of the original NTPT production methods that led to the decisions for process optimization and guided the work on the advancement of more complex hybrid-ply prepreg manufacturing techniques.

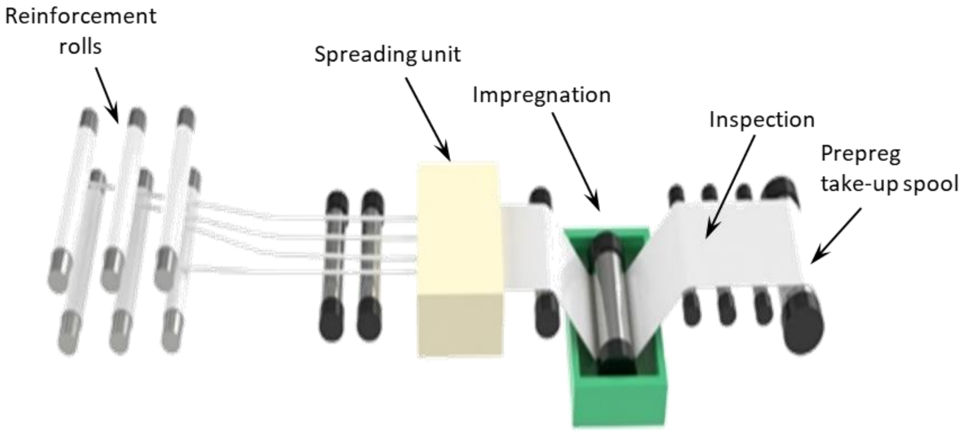


Figure 4.1 A typical prepreg line for thin prepreg manufacturing.

4.1.1 Fiber tow preparation and feeding stage

To ensure the successful production of thin-ply prepregs, the proper preparation of the raw materials (fiber and resin systems) and the processing equipment is of great importance. It is recommended that all the materials are stored in a dry area with ambient temperature and relative humidity, optimally between 20 to 25 ^{o}C and 50-70% respectively and that a first-in-first-out inventory management system is implemented to minimize potential negative effects of storage conditions. Furthermore, the fibers should be conditioned in the production environment for a minimum of 24h to minimize any impact that environmental factors may have on them. Finally, all equipment-related preparations, such as switching on the heating elements, are done at this stage, for the production line to be fully operational.

The first apparatus utilized in the process is the fiber feeding stage which typically consists of a fiber creel stand. The bobbins with the tows of fibers wound on them are loaded onto the creel stand, which is designed to hold them at a specific arrangement and control the unwinding and distribution to the subsequent stages of the prepreg production line through a series of rollers, guides, and tensioning devices. The fiber feeding stage plays a crucial role in maintaining the appropriate tension on the tows of fibers as they are guided through the process, which is essential for the proper alignment and homogeneous impregnation with the resin system.

The tension can be adjusted manually or automatically, depending on the type of creel stand and the intended application. Additionally, the fiber feeding stage/creel stand is responsible for maintaining the appropriate flow rate of the fibers into the prepreg manufacturing process. This is crucial to ensure that the fibers are fed into the process at a controlled and consistent rate, thereby preventing any quality issues in the final product, such as uneven resin content or poor fiber alignment.

4.1.2 Fiber spreading stage

The second phase in the prepreg production process is the main/primary spreading stage, during which large tows of fiber are disassembled into smaller bundles or, ideally, individual filaments. North Thin Ply Technology utilizes a proprietary fiber spreading method, thus technical details regarding the exact method will not be discussed in this work due to confidentiality concerns. However, it can be assumed that the method combines characteristics of the known methods presented in the state-of-the-art (mechanical, pneumatic, electrostatic) to offer energy to the tows of fibers and increase their width into flat unidirectional (UD) tapes.

4.1.3 Impregnation stage

The third critical stage of industrial prepreg manufacturing is the impregnation stage. This stage involves mixing and/or activating the resin system and all the processing steps required to introduce and evenly distribute it to the spread tows of fibers to fabricate unidirectional prepreg tapes of reduced thickness (thin ply prepregs). Depending on the type of resin which may come in a liquid, a viscous material, or a thin film form, the reinforcement fibers pass through resin baths, spraying nozzles, and film applicators with force the resin into the reinforcement material typically with the assistance of a set of squeezing rollers. Each method has its advantages and disadvantages, and the choice of them depends on the specific properties that each manufacturer wants to give to the final product.

North Thin Ply Technology uses a proprietary impregnation stage that evenly distributes the resin through-out the spread tows of reinforcement fibers and fully saturates and binds them to homogeneous thin-ply pre-preg. Although the exact impregnation steps are confidential, we can assume that are employ impregnation methods reported in the literature that have been specially optimized for thin-ply prepreg manufacturing.

4.1.4 Thin prepreg edge trimming

Trimming the edges of the prepregs is a common step in the manufacturing process of thin-ply prepregs. The prepreg is either carried over one or sandwiched between two sheets of silicon paper that act as a backing paper and facilitate the handling of the material. This backing paper is typically wider than the final desired width to include any prepreg width variations that may occur during production. At this stage an automated prepreg trimming method is employed, the mechanism consists of a set of rotary blades that cut the thin pre-preg to a preset width as it passes between them as depicted in Figure 4.2. The production of prepreg tapes with clear edges is essential for the elimination of common manufacturing defects such as overlaps and misalignments that can happen during lamination, especially when the material is handled with Automated Tape Laying methods (ATL), which are more sensitive to width variations of the prepreg tape. For hybrid prepregs, the quality of the borders has even greater importance since it can influence the hybrid pattern, especially for tow-by-tow configurations.

Figure 4.2 Trimming to final dimensions of the tow-by-tow carbon-glass thin ply prepreg.

4.1.5 Thin prepreg visual inspection for online quality control

Visual inspection serves as the first quality control measure in the production of thin-ply prepregs. Momentary production instabilities, unwanted variations of the process parameters, and inconsistencies between different raw material batches can result in defects and distortions within the thin-ply prepreg pattern, which can negatively impact the overall quality and mechanical properties of the final thin-ply composite part. To promptly detect defects, the thin prepregs tape is inspected by passing in front of a light table at the final stage of the production process, as presented in Figure 4.3. This simple inline quality control method allows for the identification of visually discernible defects. Figure 4.4illustrates some of the most common defects that occur during thin prepreg manufacturing: fiber or tow break, tow misalignments, twists or width variations, entrapment of foreign bodies, and poorly impregnated areas. Addressing these defects involves conducting online adjustments of the production process, to stabilize the production and to ensure that the final product conforms to the desired specifications. After this stage, and provided the prepreg passes this quality control, the material is guided to the take-up sloop, where it is rolled onto cardboard bobbins, and it is ready for immediate use or vacuum-sealed and stored in the freezer.

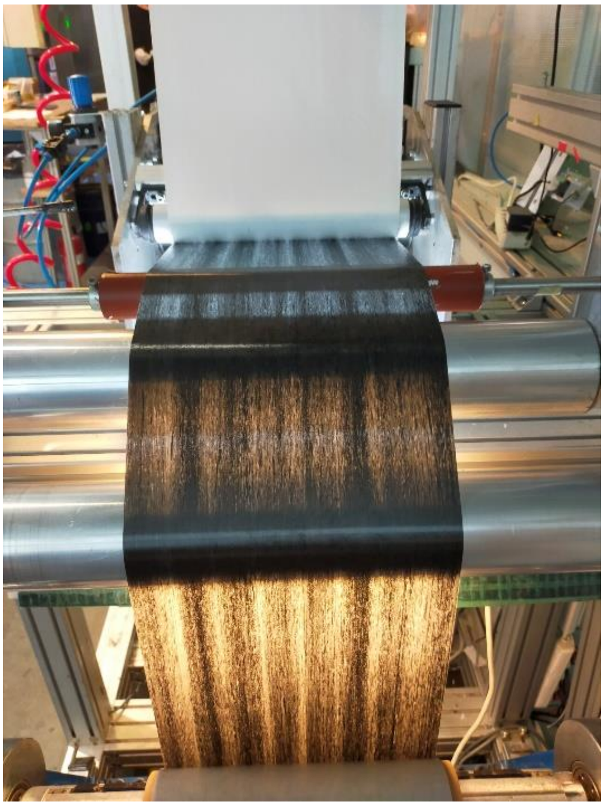


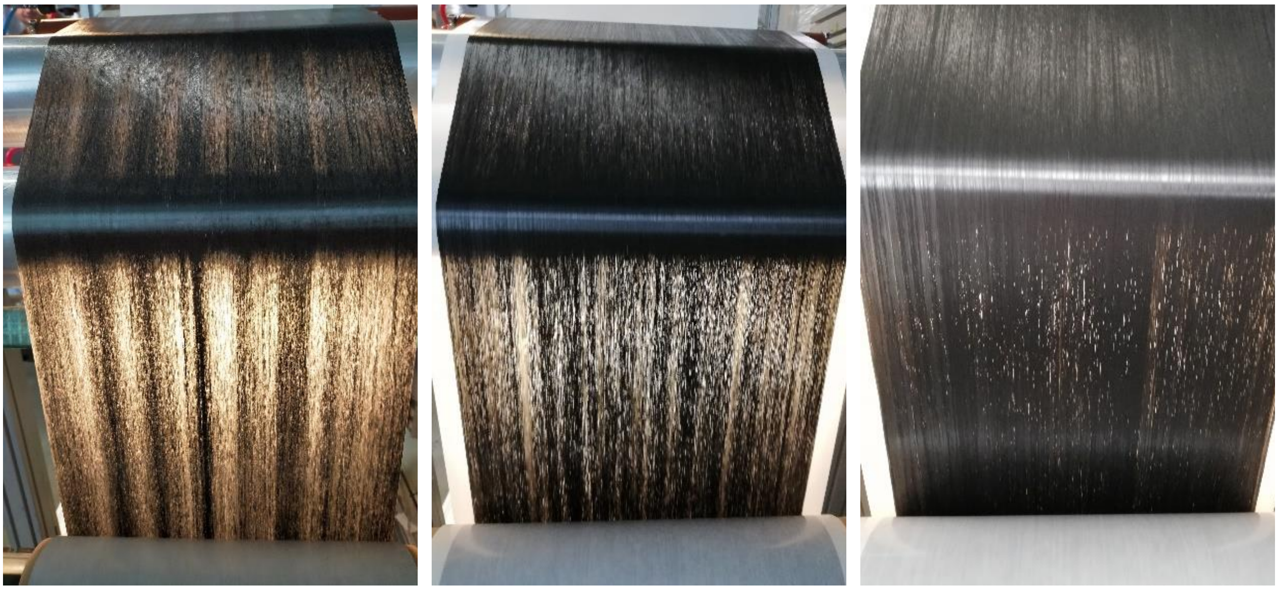
Figure 4.3 Visual inspection of thin carbon prepreg at the final stage of the production.

a b c

Figure 4.4 Prepreg defects detected with the optical inspection stage at the end of the prepreg production line. a) tow misalignments. b) tow width variations c) fiber gaps and misalignments.

4.1.6 Non-hybrid carbon fiber thin-ply prepreg production

The conventional single fiber (non-hybrid) thin-ply prepreg manufacturing method of NTPT was employed for the production of three different prepreg rolls that were used to determine the baseline properties of the HS: high strain 34-700 in two different thicknesses and the LS: low strain HR40. These prepregs were also used to prepare all the ply-by-ply hybrid configurations, as presented in G.Broggi's thesis [77]. Figure 4.5 illustrates the three different baseline prepreg as they pass in front of the light source of the visual inspection stage at the end of the production before they are rolled on 300 mm wide rolls. For the low grammage prepregs, it is visible that the tow spreading is not fully achieved, and some fiber clusters are thus expected.



a b c

Figure 4.5 a) 15 gsm HR40 b) 30 gsm 34-700 12K c) 60 gsm 34-700 24K.

4.2 Modifications to the prepreg production line and effect on fiber damage

Prepreg manufacturing methods have been reported to affect the properties of individual fibers which can later affect the quality of the prepreg. Fiber damage during prepreg manufacturing is a well-known issue in the composite manufacturing industry. During the manufacturing process, these fibers can become damaged due to various factors such as improper handling, excessive tension, or exposure to high temperatures and chemicals. Fiber damage can take various forms, such as breaking and surface cracking, as illustrated in Figure 4.6.In addition to reducing the strength and stiffness of the final composite material, fiber damage can also lead to increased production costs and decreased product quality. Several possible strategies can be employed to prevent or minimize fiber damage during prepreg manufacturing. If the fiber quality cannot be controlled, minimizing the interaction of the fiber with the spreading and impregnation stages can lead to fiber damage reduction. Minimizing the fiber/machine interaction becomes more important when developing hybridization techniques where a combination of fibers that have different physical and chemical characteristics can interact differently with the spreading mechanisms leading to uneven fiber mixing and poor quality of the final product. This study focuses on monitoring the influence of different processing routes and parameters on the properties of individual fibers and attempt to get a deeper understanding of the effects that may have on the overall quality of single-fiber E-glass/epoxy thin-ply composite materials. The results will guide the development of fiber spreading and impregnation methods for fiber hybridization.

Figure 4.6 Fiber break of a tow of carbon fiber due to friction between the filaments and the static aluminum spreading bars of the mechanical pre-spreading stage.

North Thin Ply Technology utilizes a modular thin-ply prepreg production line that can be configured to meet specific production plans and desired prepreg characteristics. Due to confidentiality restrictions though, the fiber spreading, and impregnation stages of the production line were deemed a "black box" and were excluded from the experimental study. The modularity of the prepreg of NTPT provided the opportunity to design an experimental prepreg manufacturing line based on the original process with the addition of production stages and custom-built pieces of equipment before the main and confidential fiber spreading and impregnation units. This new experimental line allowed the examination of the impact of two crucial stages of the prepreg manufacturing processes namely fiber mechanical pre-spreading and fiber thermal treatment on the quality of the fibers and the properties of the cured composite laminate. Figure 4.7 illustrates the modifications brought to the line, by addition of a pre-spreading unit, and of a thermal treatment unit.

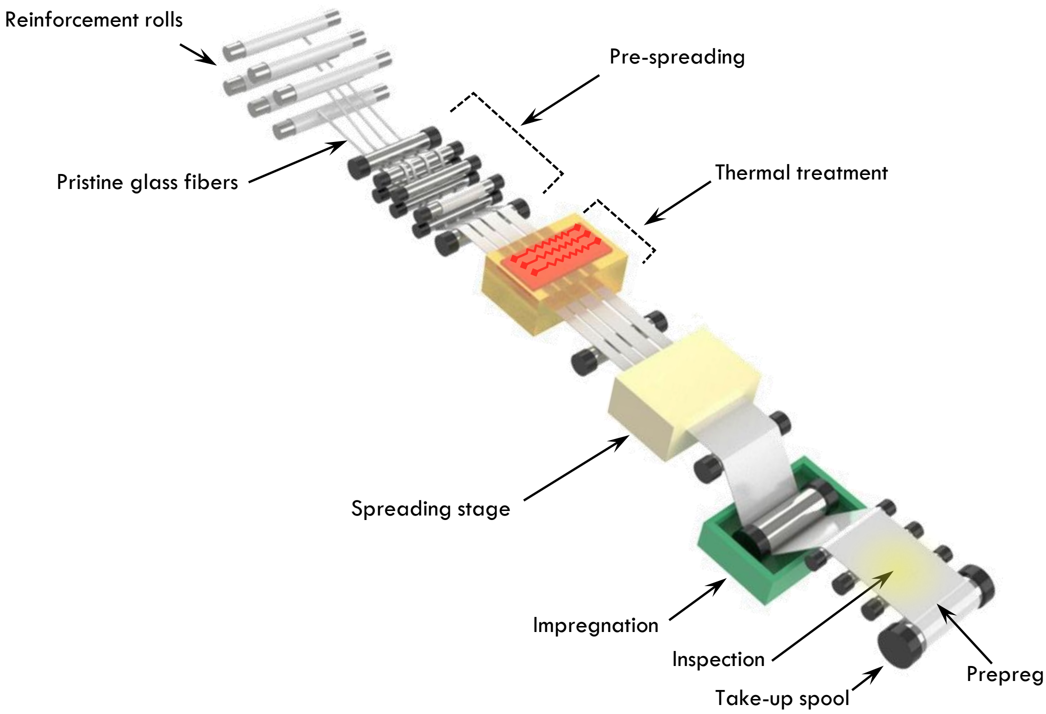


Figure 4.7 Modified fiber spreading and impregnation line employed for the manufacturing of non-hybrid thin-ply prepregs.

The following sections will discuss in detail how the new production stages were integrated into the original prepreg line, their working principles, and their role in the hybrid thin-ply prepreg production.

4.2.1 Mechanical pre-spreading stage

This new mechanical fiber pre-spreading stage was designed in addition to the original NTPT fiber spreading and impregnation line, having in mind that any performance improvement could be easily adapted to produce more complex hybrid thin-ply materials. The concept idea was that the initiation of the tow spreading before the main spreading zone of the NTPT prepreger can bring significant improvements to the production process. Firstly, the pre-spreading stage can ensure a gradual disassembly of the large tow into smaller sections which can later enter more efficiently the main spreading stage. This even distribution of the spreading steps can promote fiber alignment and spreading uniformity. Secondly, the pre-spreading stage can also bring improvements to the impregnation quality as evenly distributed fibers have more exposed surface area and can be saturated with the resin system more effectively compared to larger bundles that require forcing the resin system into them. These improvements can have a direct positive impact on the quality of the prepregs as increased fiber alignment and even resin distribution could enhance the mechanical properties of the composite materials, lead to a reduction in processing time, thus increasing overall production efficiency which can result in cost savings over time. The custom-made fiber pre-spreading production stage developed for this study consists of an aluminum frame on which multiple static rods or rollers can be positioned in the required configurations, as presented Figure 4.8.

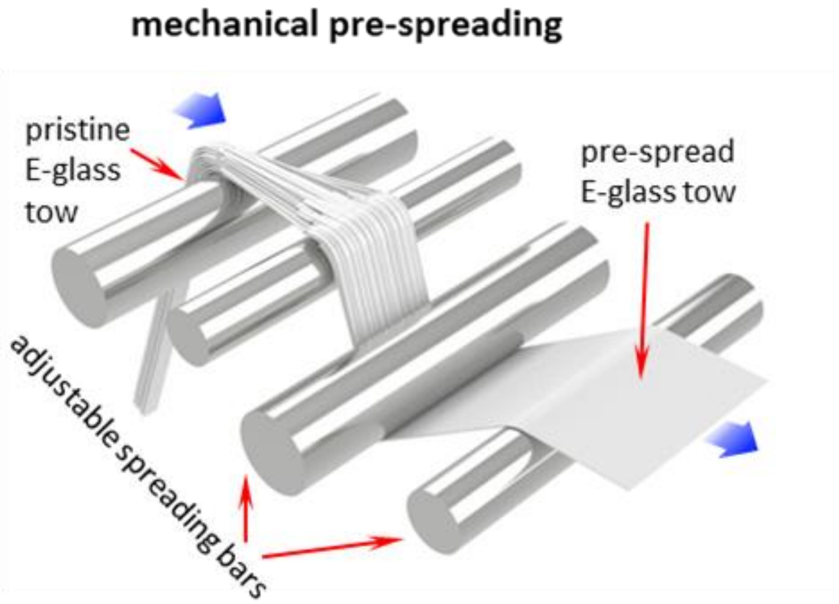


Figure 4.8 Schematic of fiber pre-spreading stage.

The fiber tows are unwound from the creels and guided through several static spreading bars on an aluminum frame. Depending on the manufacturing requirements and the level of spreading needed for each prepreg, several parameters can be adapted: number of bars, diameter, angles, and distance between them.

The optimal pre-spreading configurations (the number of rods, angles, and distances between them) were determined for each type of fiber separately with several test runs, evaluating visually and with tow width measurements the quality of the spreading. This ensured maximum pre-spreading with minimum fiber damage due to contact friction between the equipment and the tows. Figure 4.9 shows a schematic of fiber pre-spreading stages.

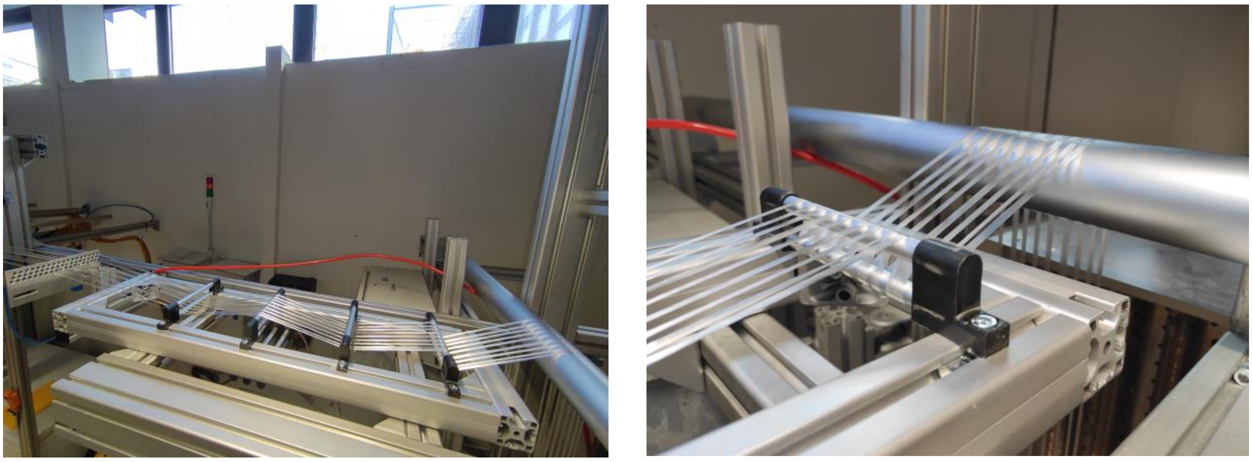


Figure 4.9 Implementation of a fiber pre-spreading stage on the modular experimental thin prepreg production line.

4.2.2 Thermal treatment stage

Commercial reinforcing fibers are generally supplied with a coating usually referred to as “sizing”. Fiber sizing usually includes coupling agents, anti-static agents, lubricants, and binding media that tend to make the individual filaments of a tow stick together yet limit fiber breaking due to the friction with the spreading equipment (roller, vibrating bars, airflows). The characteristics and relative concentration of the sizing present on the surface of the filament both have significant importance for thin ply prepreg manufacturing as they strongly influence the ability of filaments in the tow to separate and spread up to the desired, single fiber level. The manufacturing of hybrid thin-ply composites, in addition requires the simultaneous handling of fibers with dissimilar sizing characteristics. Therefore, a sizing removal step was added to the process to start from a common basis and eliminate the influence of this parameter. Before entering the main spreading and impregnation units, the tows are passed through a hot chamber where an array of infrared (IR) lamps partially degrades the chemical compounds present on the surface of each filament, as illustrated in Figure 4.10. The parameters to adjust in this stage are the time within the chamber, and the power of the lamp.

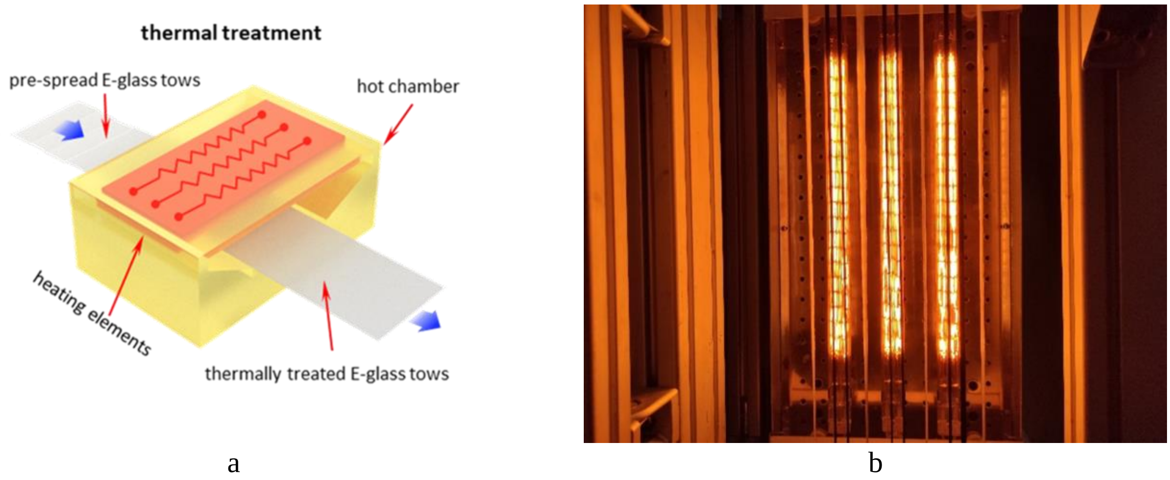


Figure 4.10 Thermal treatment stage a) Schematic of fiber coating removal stage b) fiber tows inside the thermal treatment chamber.

4.3 Effect of thin-ply prepreg manufacturing processing on the fiber and composite strength

This section presents the experimental campaign aiming to identify the effect of the modified processing stages as described and optimized previously, on the strength of single E-glass fibers and then on the strength of E-glass/epoxy (TP415) thin-ply composite materials. This section is based on the Master thesis work of Yannick Aubry performed at EPFL, and at Technical University of Denmark (DTU) in collaboration with HyFiSyn partners Dr. Rajnish Kumar, Dr. Lars P Mikkelsen, and on the publication in preparation: Effect of thin-ply prepreg manufacturing processing on the fiber and composite strength - Rajnish Kumar, Alexios Argyropoulos, Yannick Aubry, Lars P Mikkelsen, Hans Lilholt, Véronique Michaud, and Bo Madsen.

The E-glass fiber treatment and the production of the thin-ply E-glass prepregs was carried out at NTPT, the production of the thin-ply E-glass/epoxy composite laminates was conducted at EPFL. Both single fibers and composite materials testing, and analysis were carried out at DTU.

Figure 4.11 presents the three main routes proposed for the processing of the prepregs: 1- Pristine corresponds to the in-house NTPT processing method, 2- Pre-spread adds the pre-spreading stage optimised for glass fibers, and 3- Pre-spread and thermally treated adds the thermal treatment stage, optimized for glass fibers.

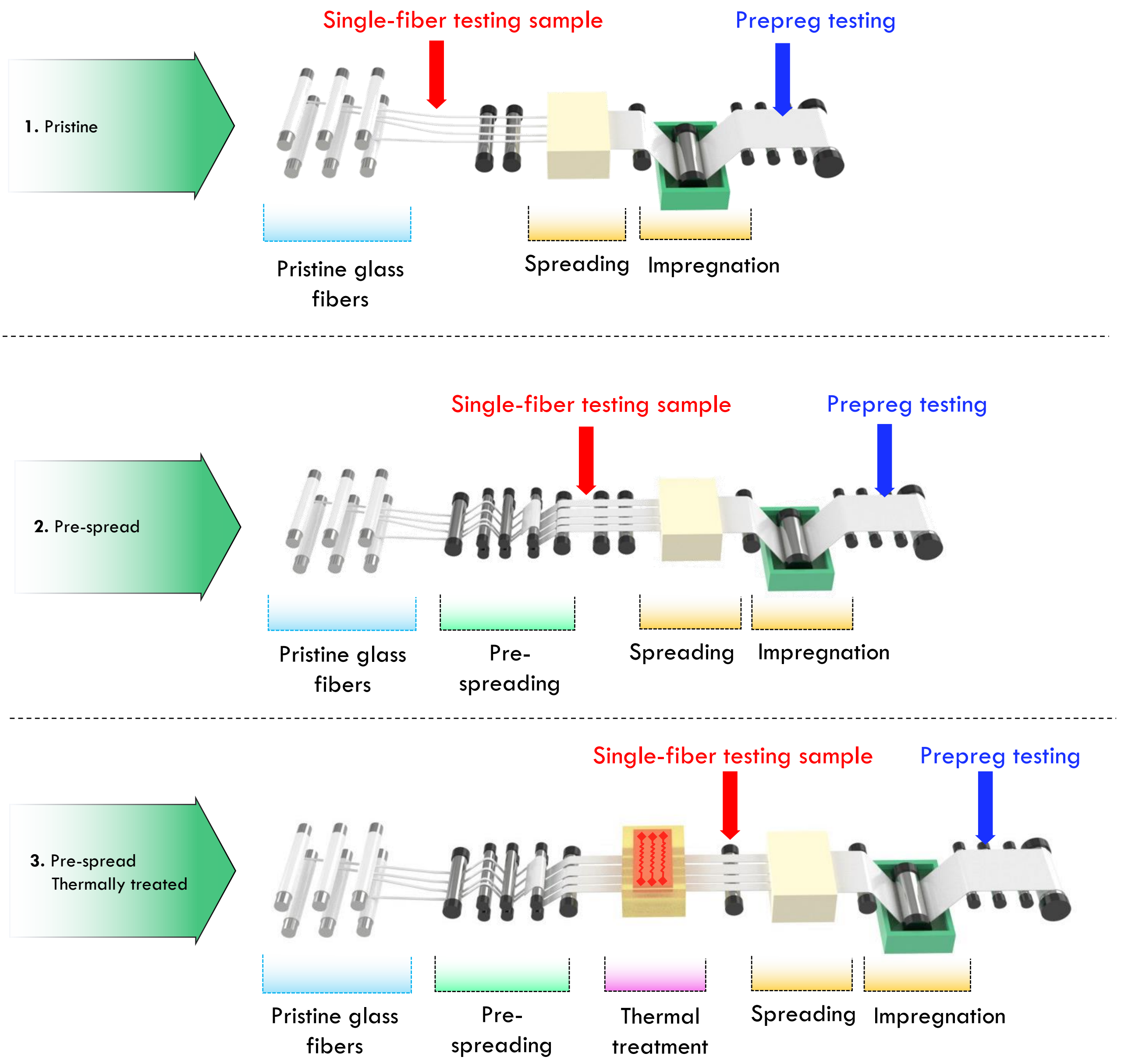
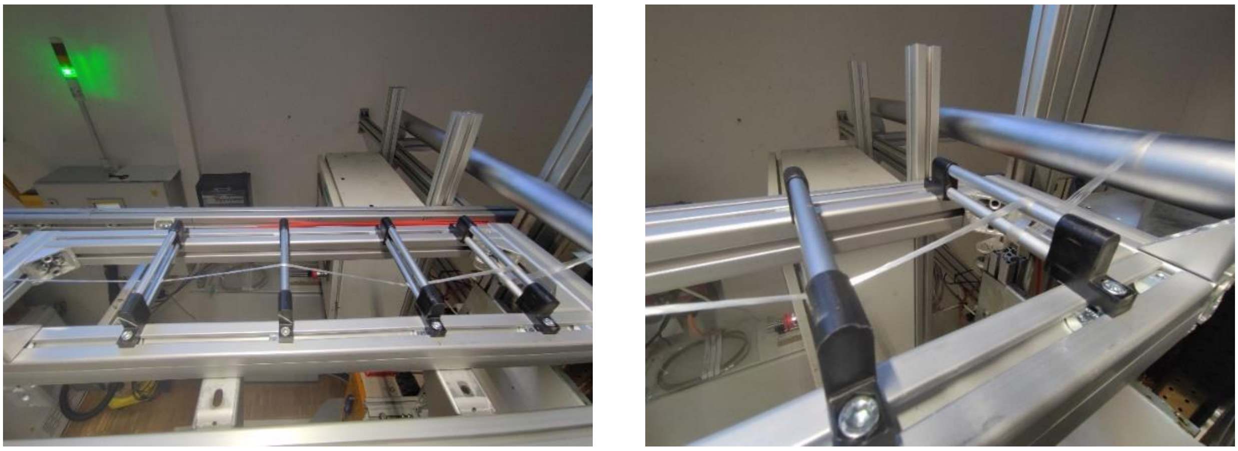


Figure 4.11 The three types of E-glass/epoxy thin ply prepregs manufactured to study the effects of different processing routes on the quality of the final production.

4.3.1 Single fiber extraction, characterization and testing methods.

Three batches of fibers were extracted for single fiber testing at the different stages of the modified thin-ply prepreg assembly, as shown in Figure 4.11. The first batch was extracted directly from the bobbin. It was used as a baseline and will be referred to as "as-received" (R). The second batch was extracted after the pre-spreading stage and will be referred to as "pre-spread" (PS). Finally, the third batch was extracted after both the pre-spreading and thermal treatment stages and will be referred to as "pre-spread and thermally treated" (PST). Figure 4.12 shows examples of the tow positioning in the line.



a b

Figure 4.12 Treating a tow of E-glass to produce specimens for single fiber testing. a) tow of E-glass fibers passing through the fiber spreading stage. b) tow of E-glass fibers entering the thermal treatment stage.

Density measurement

The density of three glass fibre types (R, PS and PST) was measured by the gas pycnometry method (Ultrapyc 1200E; Quantachrome Instruments, USA). Two samples were measured for each fiber type, with 100 runs for each sample. The measured density values are presented in Table 4-1. No significant difference in the fiber density measurements is observed between the fiber types.

Table 4-1 Fiber density measurements ρ_f

	As-received (R)	Pre-spread (PS)	Pre-spread + thermal conditioned (PST)
Fiber density [g/cm^3]	2.6270 ± 0.0001	2.6250 ± 0.001	2.6250 ± 0.0003

Single fiber tensile tests:

Single fiber tensile tests of the three glass fiber types (R, PS and PST) were performed using a semi-automated single fiber testing machine “Favimat+” combined with Robot 2 (TexTechno, Mönchengladbach, Germany), with a built-in vibroscope system to determine the fiber cross-sectional area. Based on the first vibroscope test, the linear density (T [kg/m]) of the fiber was determined using a recently developed iterative analysis [114], and the cross-sectional area (A=T/ρ) of the fibre was then calculated using the measured fibre density (ρ [kg/m^3]). Fibres were tested at five-gauge lengths (15, 20, 30, 40, and 50) mm, using 150 fibers for each gauge length. Measuring the apparent fibre modulus for each gauge length, a gauge length dependent stiffness is observed. From this dependency, the compliance (C) was determined [115]. All the recorded displacement was subsequently corrected for this compliance removing this unphysical gauge length dependent stiffness. The test speed (clamp speed) was 1 mm/min. The approximate tensile testing time for each fibre is 1-2 minutes. A video describing the machine’s set-up is available in [116].

The two-parameter Weibull model [117] was used to analyse the glass fibre's failure probability statistically. In the Weibull model, the probability of failure P_F of a material at a given stress σ and with a given length L can be written as:

$$P_F(\sigma, L) = 1 - \exp\left(-\frac{L}{L_0}\left(\frac{\sigma}{\sigma_{0(L_0)}}\right)^m\right) \tag{4.1}$$

where m is the Weibull modulus, L_0 is a reference length, and $\sigma_{0(L_0)}$ is the so-called characteristic strength at a failure probability of 0.6321 at the reference length.

4.3.2 Composite manufacturing, characterization and testing methods

Two types of E-glass/epoxy thin-ply prepregs were manufactured, one with pre-spread tows (PS) and the other using pre-spread and thermally treated tows (PST). We targeted a fiber areal weight (FAW) of 50 g/m^2 (gsm) and a nominal 38% resin content (RC) by weight. An equivalent NTPT prepreg was also used as reference for comparison of the fiber orientation. The thin prepregs were stacked manually to form multi-layer unidirectional (UD) 0^0 laminates. To reach a cured plate thickness of over 2 mm that meets the requirements of the mechanical testing standards laminate, preforms were produced by stacking 64 thin-ply prepreg layers. The preforms were trimmed to 700x500 mm to create large laminates and then cured in an autoclave with the following the process that has been presented in Chapter 3.

Fiber volume fraction

The fiber volume fraction (V_f) of prepreg-based composite specimens manufactured with PS and PST rovings was determined using the gravimetric method (ASTM D3171-15, ASTM D2584). Five samples with a rectangular area of 15×15 mm^2 were cut from the composite laminates. The density of the samples was measured using the buoyancy method based on Archimedes's principle. The fiber weight fraction of the samples was determined by the resin burn-off method. The fiber volume fraction of the composite was calculated by standard equations [5] using densities of glass fibers and matrix. The V_f of composite specimens manufactured with PS and PST rovings were determined to be 62.0 ± 1.0 % and 59.3 ± 1.4 %, respectively. An alternative way to detect the fiber volume fraction is based on an image analysis of high-resolution scanning electron microscopy (SEM) images of the full cross section similar to what is done in [118].

Fiber orientation

The fiber orientation of composite specimens manufactured with PS and PST rovings was characterized by the non-destructive X-ray Computed Tomography CT technique using ZEISS Xradia 410 Versa microscope. Structure tensor analysis was then used for orientation characterization.

The fiber orientation of composite specimens manufactured with pre-spread and pre-spread + thermally conditioned fiber rovings was characterized by the non-destructive X-ray Computed Tomography CT technique using ZEISS Xradia 410 Versa microscope. Structure tensor analysis was then used for orientation characterization [6].

Static tensile testing in the longitudinal direction

The test specimen geometry used in this study was a non-straight-sided geometry called X-butterfly, shown in Figure 4.13 [7], [8]. Press-molded woven prepreg biaxial glass composite laminate (Electro-Isola A/S, Vejle, Denmark) with a thickness of 2 mm was used as end-tab material, with the fibers positioned at ± 45^o to the longitudinal direction. The surfaces of the specimens at the end-tab areas were slightly ground using a disc sander and were cleaned with ethanol. The end tabs were bonded to the specimens using a structural epoxy adhesive (3M, DP460) with a cure cycle of room temperature for 24 h and 40 C for 16 h. The X-butterfly test specimens were machined using a CNC milling machine.

The tensile test was performed on a universal testing machine (Instron 88R1331, Norwood, MA, USA) with a 50 kN load cell. The crosshead speed was 2 mm/min for all specimens. The strain was measured with two 6 mm long single strain gauges (LY11-6/350) placed on the center on each side of the test specimens. Six specimens of each specimen type manufactured with PS and PST rovings were tested.

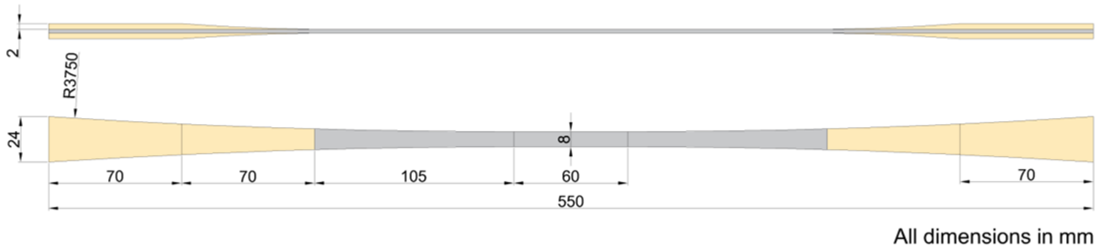


Figure 4.13 Geometry and dimensions of X-butterfly test specimens.

Static tensile testing in the transverse direction

The test specimen geometry for the tensile tests in the transverse direction is shown in Figure 4.14. The test specimens were machined using a CNC milling machine. The tensile tests were performed on a universal testing machine (Instron 88R1331, Norwood, MA, USA) with a 50 kN load cell using mechanical grips with a load limit of 5 kN. Extensometers with a gauge length of 12.5 mm were used for strain measurement. The modulus measurements were performed in the strain range of 0.05 -0.25 %. Six specimens of each specimen type manufactured with PS and PST rovings were tested.

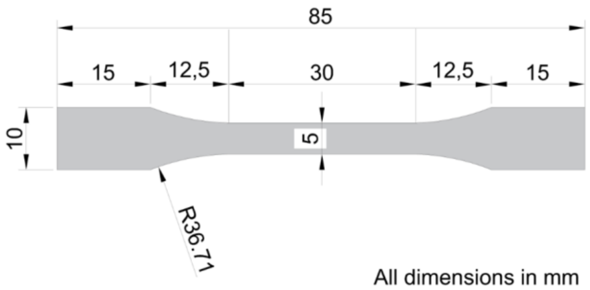
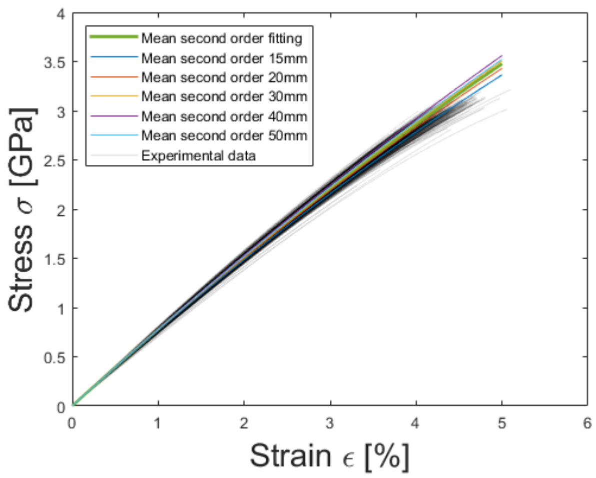


Figure 4.14 Geometry and dimensions of transverse tensile test specimens

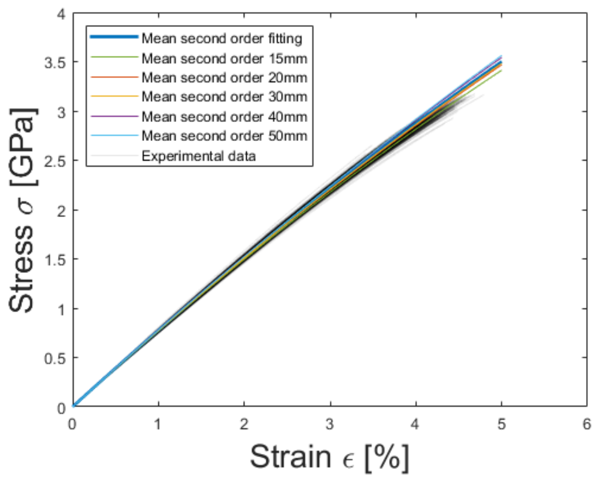
4.3.3 Results and discussion on single fiber testing

Modulus measurement

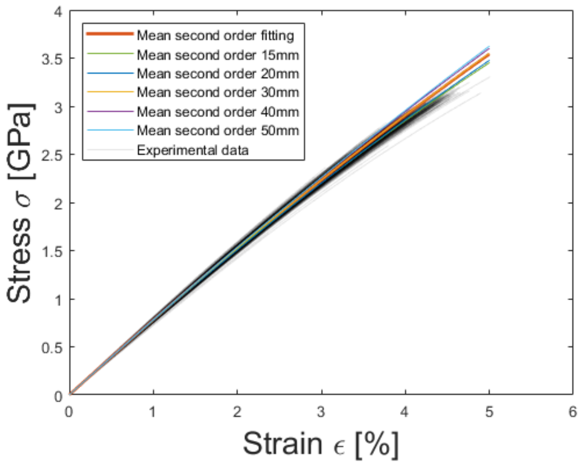
The single fiber stress-strain curves are presented in Figure 4.15



(a)



(b)



(c)

Figure 4.15 Experimental stress-strain curves for single fibers. a) as received, R, b) pre-spread, PS, c) pre-spread and thermally conditioned, PST

The measured modulus (compliance corrected) of R, PS and PST single glass fibers are summarized in Table 4-2. The modulus values for the tested fiber types (R, PS, and PST) showed almost similar values within the standard deviation. This confirms that the different fiber treatment steps did not influence the fiber modulus.

Table 4-2 Measured fiber modulus for single glass fibers (as received, pre-spread and pre-spread + thermally conditioned).

	As-received (R)	Pre-spread (PS)	Pre-spread + thermal conditioned (PST)
Fibre modulus [GPa]	77.6 ± 1.5	77.5 ± 1.2	78.0 ± 1.6

It was however noticed that the curves followed the shape of a second-order polynomial curve, as reported in [119]. The curves were thus also fitted with the following equation: $\sigma(\varepsilon)=1/2\ \alpha\ \varepsilon^2+E_0\ \varepsilon$, where E_0 is the initial stiffness at ε =0, and α [MPa] is a coefficient. These fits will subsequently be used in the strength model later on, with E_0=78 GPa, and α =-354.5 MPa as an average of all fits.

Statistical analysis

The Weibull plots of R and PS glass fibers are presented Figure 4.16 and Figure 4.17, and show a slight non-linear trend with an R-squared value for a linear regression fit of 0.93 and 0.94, respectively. Non-linear Weibull distribution has often been observed for glass fibers and has been related to multiple failure modes associated with different types of the flaw population in the material [120], [121]. Yang et al. [122] proposed to use a bimodal Weibull distribution to analyze the strength distribution of sized glass fibers with a reasonably good fit. They hypothesized that two exclusive types of flaw populations might control the failure of the sized fibers. One flaw type corresponds to defects found on the bare surface of the glass fiber, and the other relates to the surface flaws on the glass surface coated with sizing, leading to a different behavior.

The Weibull plots of PST glass fibers on the other hand show a linear trend with an R-squared value for a linear regression fit of 0.98. Yang et al. [122] also characterized commercially manufactured bare (un-sized) single glass fibers. They observed that the Weibull plots for the un-sized fiber show a linear trend at each gauge length, implying that a single-flaw population dominates the failure. The linear trend of the Weibull plots of the PST fibers suggests that the thermal conditioning treatment potentially led to the partial removal of the sizing. This phenomenon was also physically observed as some fumes were observed during the thermal treatment stage, i.e. passing the fibre bundles through the heating elements.

All Weibull model parameters (m and $\sigma_{0(L)}$) for single glass fibres (as received, pre-spread and pre-spread+thermally conditioned) tested with the five-gauge lengths L $\in$ [15; 50] mm are reported in Table 4-3. Overall, the values found follow the same trends as found in the literature, with a $\sigma_{0(L)}$ value between 2 and 3 GPa. In particular, Mesquita et al. [123], using the same fiber and an automated testing device, found an average $\sigma_{0(L)} = 2.93\ GPa$. and m =5.61. The values of m found in that and our study are overall rather high, denoting that the automated testing device also prevents further fiber damage due to handling operations.

The effect of the pre-spreading and thermal treatment stages on the fibers has been analyzed by comparing the Weibull characteristic strength $\sigma_{0(L)}$ as a function of the tested gauge length of R, PS and PST fibers, plotted on Figure 4.19. The R fibers were found to have the highest strength values. Selecting the linear fit of strength values of R fibers as the baseline, the PS fibers globally showed lower strength values; this trend becomes more significant with increasing gauge lengths. This trend suggests that passing fiber tows through several spreading bars under tension during the pre-spreading stage leads to some damage to the fibers, thus increasing the defect population, resulting in decreased strength values. Compared to R and PS fibers, the PST fibers showed the lowest strength values. This trend suggests that the combined effect of pre-spreading and thermal treatment stages has further reduced the strength of the fibers. The thermal treatment stage in this study potentially leads to the sizing's partial removal. Several researchers [120], [124], [125] have reported that the surface flaws tend to head when sizing is applied. The sizing fills the severe flaws with a three-dimensional network of the silane-coupling agent, which forms covalent bonds with the glass and thereby effectively reduces the depth of the surface flaw. Feih et al. [126] studied the strength degradation of glass fibers at high temperatures. They found that the reduction in strength could be attributed to the growth of pre-existing surface flaws or the creation of new flaws during the heat-treatment process.

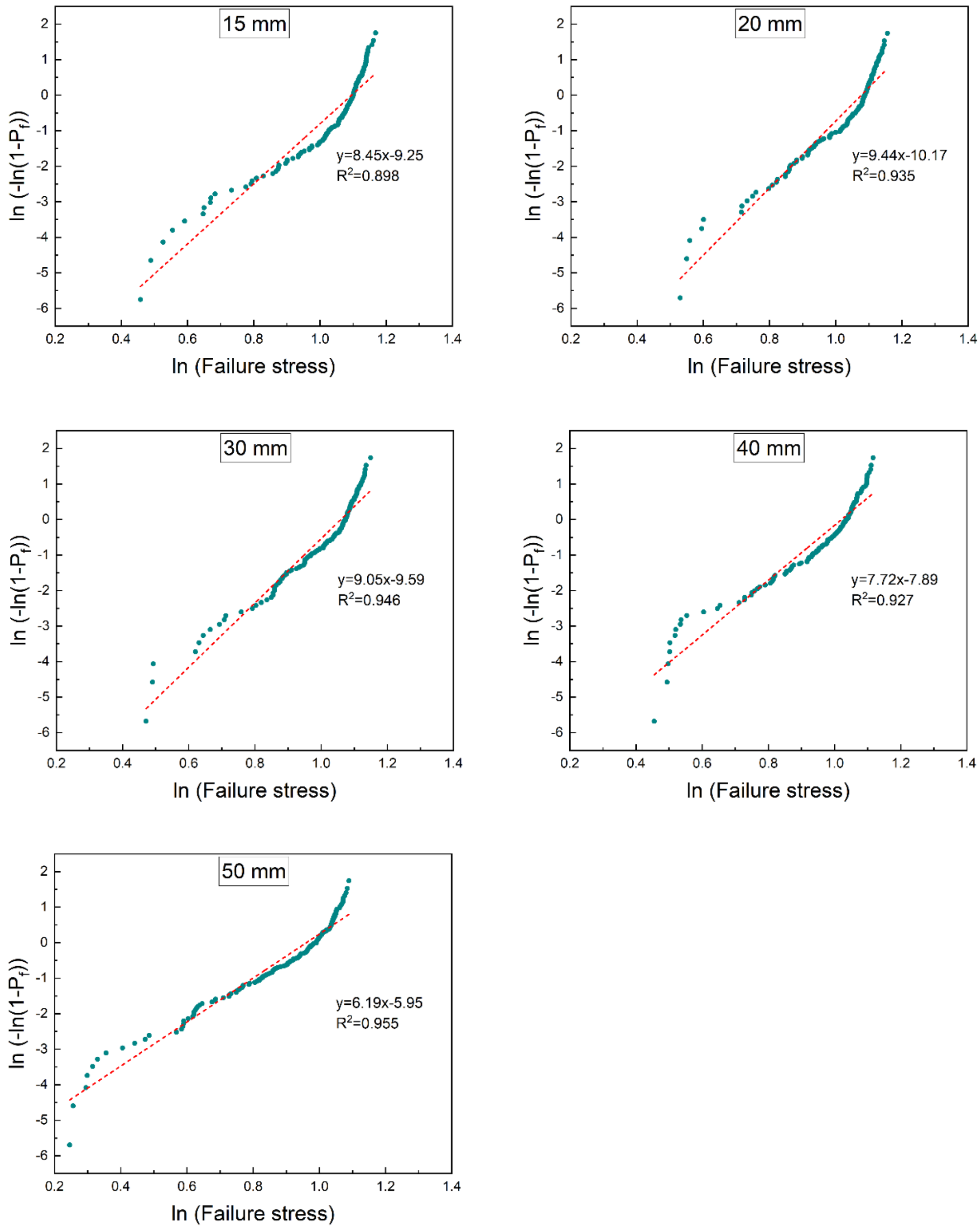


Figure 4.16 Individual Weibull plot of failure stress of as-received (R) single glass fibres for five-gauge lengths L ∈ [15; 50] mm. The dashed lines represent linear fit lines.

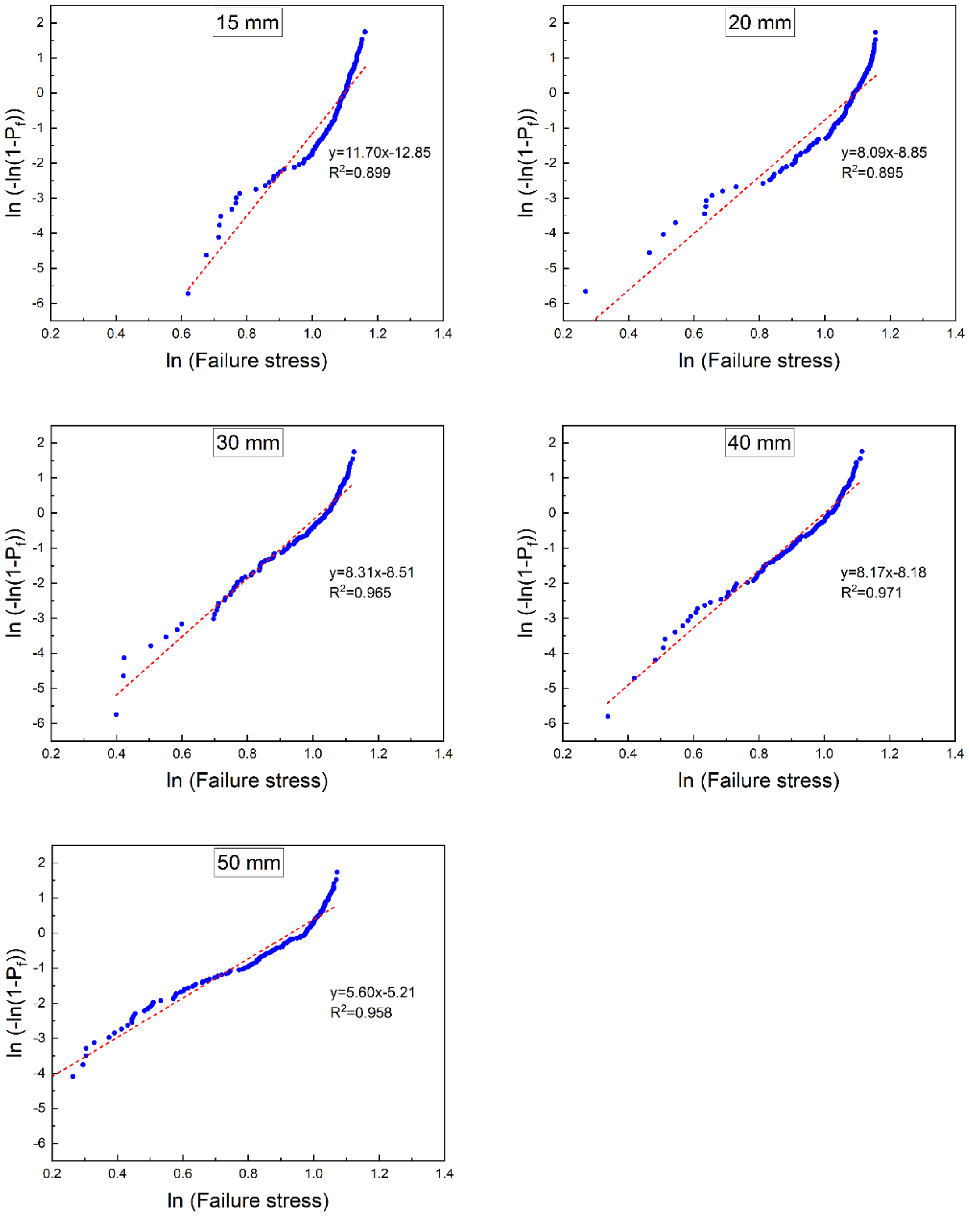


Figure 4.17 Individual Weibull plot of failure stress of pre-spread (PS) single glass fibres for five-gauge lengths L ∈ [15; 50] mm. The dashed lines represent linear fit lines.

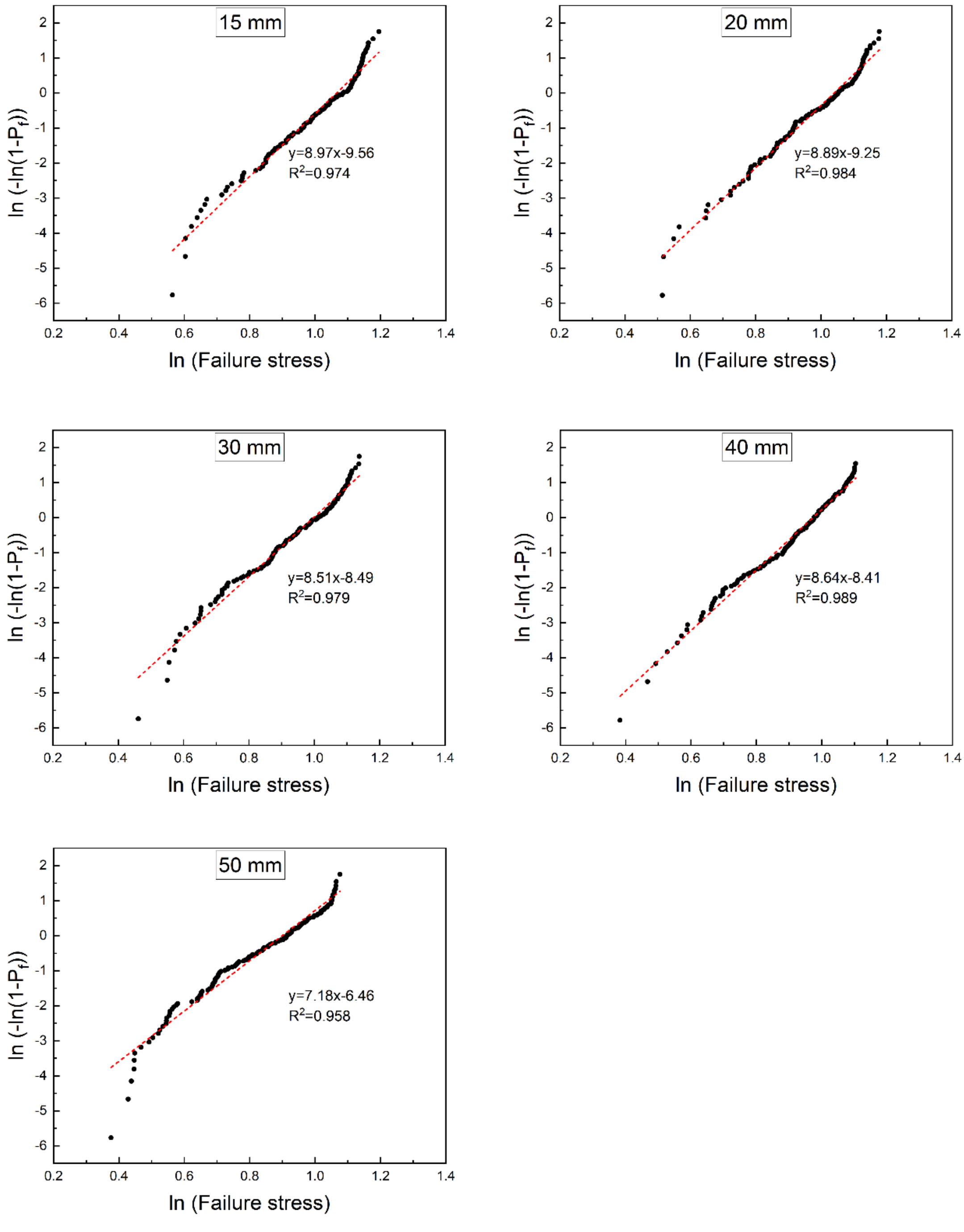


Figure 4.18 Individual Weibull plot of failure stress of pre-spread + thermally conditioned (PST) single glass fibres for five-gauge lengths L ∈ [15; 50] mm. The dashed lines represent linear fit lines.

Table 4-3 Determined Weibull model parameters (m and $\sigma_{0(L)}$) for single glass fibres (as received, pre-spread and pre-spread+thermally conditioned) tested with five-gauge lengths L ∈ [15; 50] mm.

	As-received (R)			Pre-spread (PS)			Pre-spread + thermally conditioned (PST)		
Gauge length [mm]	$\sigma_{0(L)}$ [GPa]	m [-]	R^2	$\sigma_{0(L)}$ [GPa]	m [-]	R^2	$\sigma_{0(L)}$ [GPa]	m [-]	R^2
15	2.99	8.45	0.898	3.00	11.70	0.900	2.90	8.97	0.974
20	2.94	9.44	0.935	2.99	8.09	0.896	2.83	8.89	0.984
30	2.89	9.05	0.946	2.78	8.31	0.966	2.71	8.51	0.979
40	2.78	7.72	0.927	2.72	8.17	0.981	2.65	8.64	0.989
50	2.61	6.19	0.955	2.54	5.60	0.959	2.46	7.18	0.959

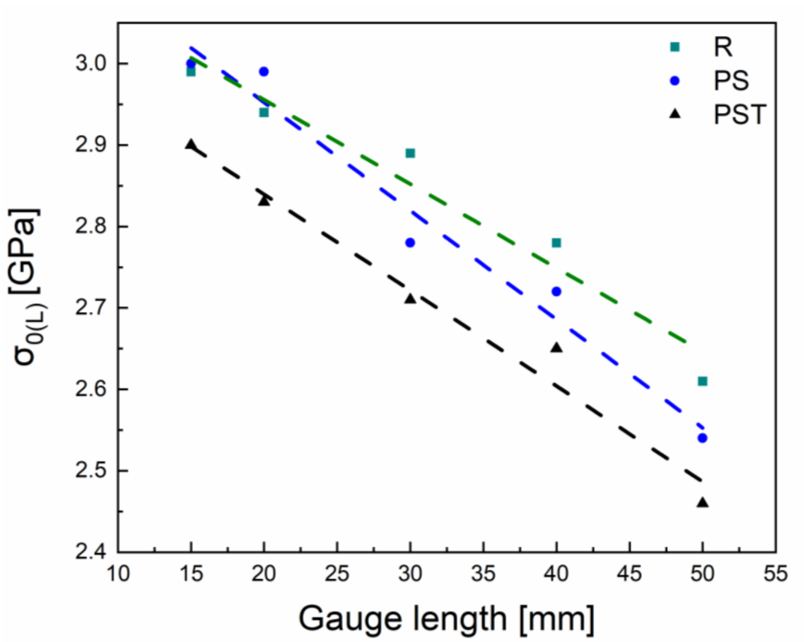


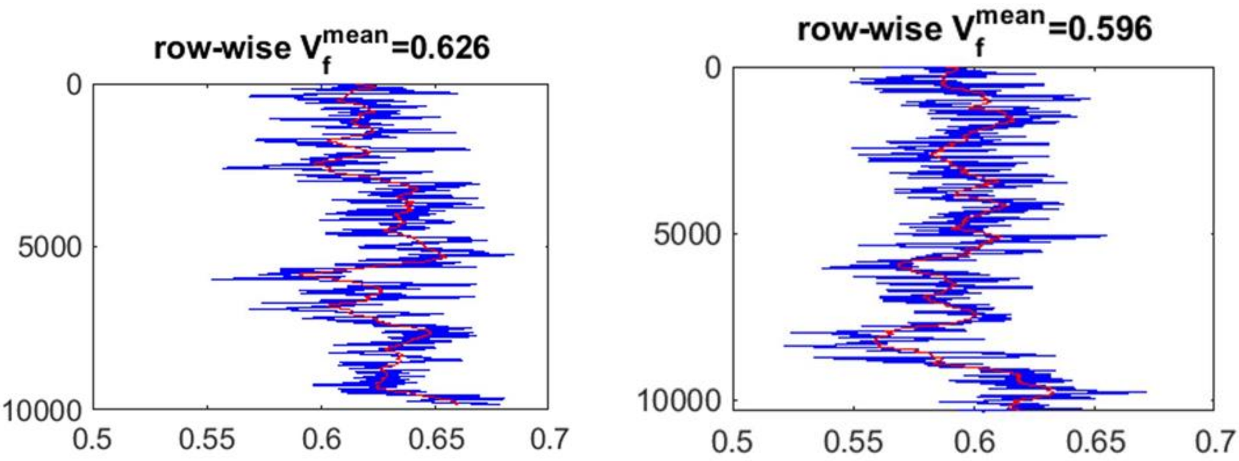
Figure 4.19 Weibull characteristic strength $\sigma_{0(L)}$ of as-received (R), pre-spread (PS) and pre-spread + thermally conditioned (PST) single glass fibres at different gauge lengths. The dashed lines represent linear fit lines.

4.3.4 Results and discussion on composite characterization and testing

The produced PS and PST composites were cut, polished, and observed by SEM, as illustrated Figure 4.20. The porosity content was very low in all samples, and they all showed very strong homogeneity, thanks to the use of 64 stacked thin plies. Figure 4.21 presents the local volume fraction analysis based on the SEM images, using an image analysis code at DTU using Otsu's method [127] , confirming the narrow fiber distribution.

The average values of V_f correspond closely to the pycnometry measurements, confirming that the PST composites had a slightly lower V_f than PS.

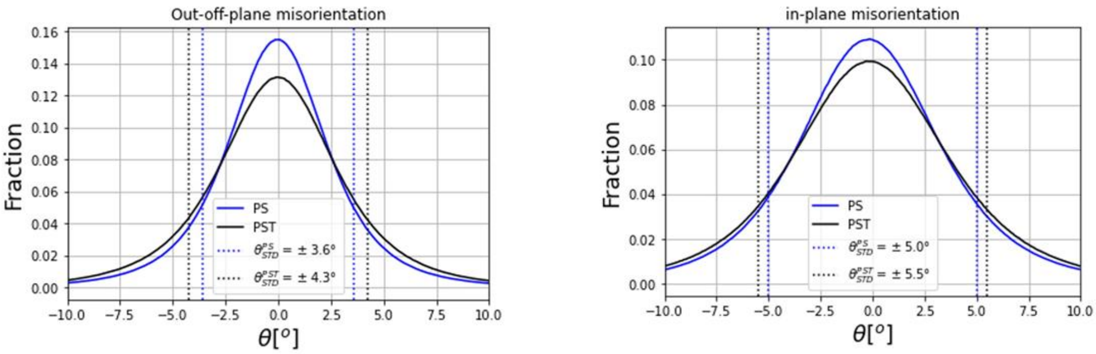
Figure 4.20 SEM image showing the microstructure of composite specimens a) PS b) PST. Thickness of the samples was 2 mm.



a b

Figure 4.21 Local fiber volume fraction measurement from SEM analysis, for a) PS and b) PST samples.

The fiber orientation was characterized using the non-destructive X-ray Computed Tomography technique, using a python code developed at DTU. In-plane and out-of-plane results are reported in Figure 4.22. The distributions curves almost overlap, and standard deviations are similar. Nonetheless, the PS composites overall show a slightly narrower distribution both in-plane and out-of-plane, indicating that the thermal treatment possibly allowed more fiber movement, resulting in a lower packing density due to the slight fiber misorientation.



a b

Figure 4.22 Out of plane (a) and in-plane (b) fiber orientation fraction, for PS and PST composites.

The results of the static tensile tests in longitudinal and transverse directions are presented in Table 4-4. Figure 4.23 presents the experimental results for the longitudinal tensile tests. As the fiber contents are slightly different, the tensile modulus can be normalized to V_f =60% (neglecting the modulus of the resin phase, which is 3 GPa according to the datasheet), as shown in the Table as well. A slight difference is observed between the samples, the PST being slightly lower in stiffness, even though the fiber modulus is similar and in strength, which is possibly expected, due to a lower fiber strength. The stiffness values for the PS composite are nonetheless close to what is expected for a UD glass fiber composite, E =47.7 GPa (for V_f=60% and a fiber modulus of 77.5 GPa as measured earlier, using the rule of mixtures).

Table 4-4 Mechanical test results in longitudinal and transverse direction for both PS and PST composites.

	Fibre volume fraction (%)		Tensile modulus [GPa]	Tensile strength [MPa]	Strain to failure [%]
Pre-spread (PS)	62.0 ± 1.0	Longitudinal	49.1 ± 0.8	1416 ± 24	3.14 ± 0.09
		Longitudinal normalized to 60%	47.5	1370	
		Transverse	11.7 ± 0.7	52 ± 2	0.57 ± 0.06
Pre-spread + thermal conditioned (PST)	59.3 ± 1.4	Longitudinal	43.9 ± 1.4	1303 ± 48	3.08 ± 0.06
		Longitudinal normalized to 60%	44.4	1318	
		Transverse	9.0 ± 0.3	47 ± 9	0.70 ± 0.20

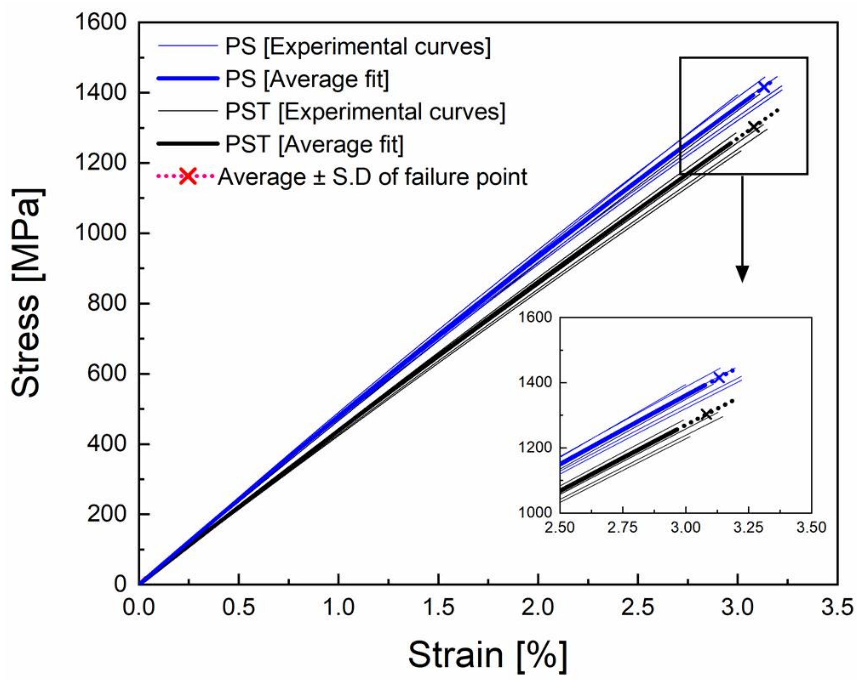


Figure 4.23 Experimental stress-strain curve for PS and PST composites in the longitudinal direction.

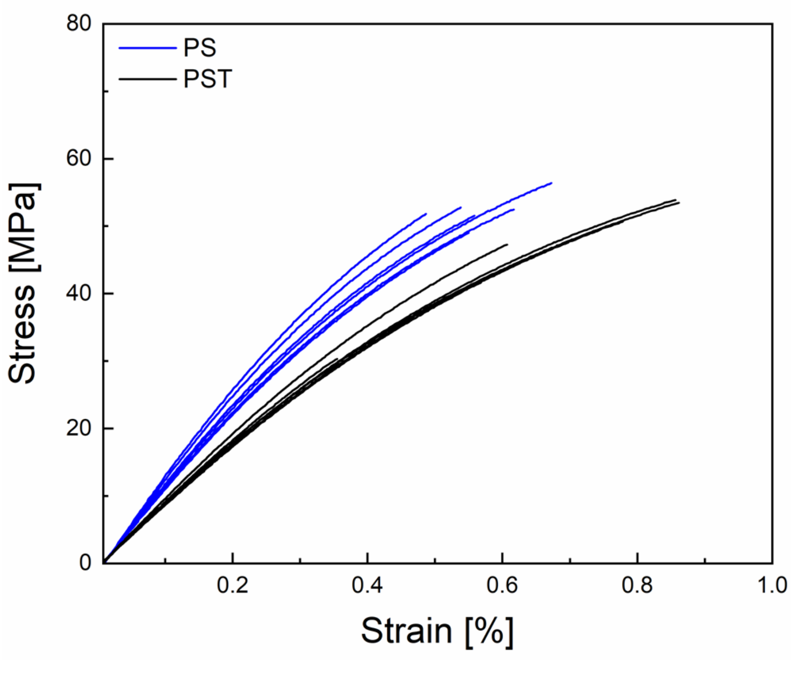


Figure 4.24 Stress-strain curve for PS and PST composites in the transverse direction.

Figure 4.24 presents the experimental results for the transverse tensile tests. Again, a slight transverse modulus difference is observed, which cannot be solely attributed to the difference in fiber content. A larger scatter is also observed for PST composites in terms of strength.

To evaluate the link between the fiber statistical failure behavior, and the composite tensile properties, the use of an analytical model based on the Global Load Sharing (GLS) model proposed by Curtin et al. [128] explored. The GLS model assumes that the matrix fails rapidly through cracking or yielding, and that fibers hence carry the load, which is redistributed through the matrix when fibers break. As presented in Chapter 2, the apparent stress in the composite is thus:

$$\sigma = V_f E_f \varepsilon - \frac{1}{2} V_f \sigma_c \left(\frac{E_f \varepsilon}{\sigma_c}\right)^{m+2} \tag{4.2}$$

where σ_c is the stress at critical pull-out length, given by:

$$\sigma_c = \left(\frac{\sigma_0^m \tau_d L_0}{r}\right)^{\frac{1}{m+1}} \tag{4.3}$$

where τ_d is the interfacial shear strength of the interface and r the fiber radius [102].

This plot is presented Figure 4.25, for both cases of PS and PST composites, using the average of the Weibull modulus for each category, as well as the average of the product of Weibull strength and gauge length, V_f =60%, E_f as a quadradic function of strain, a fiber radius of r =7.62 µm, and assuming an interfacial strength τ_d = 20 MPa.

The results show a good agreement of the predicted composite modulus but overestimate the composite strength in both cases by about 30%, which has also been reported by other authors [102]. A parametric study of the influence of the interfacial shear strength value showed little effect on the prediction, only a slight increase with a shear strength increase of 0.3 MPa in the maximum value, within the range of interest (for an interfacial shear strength between 10 and 50 MPa). The characteristic strength σ_0 has a linear influence on the composite strength value, and m has a very strong influence on the composite strength value, as expected, in particular for low values of m which led to a strong increase in the composite strength. In our case, the values of m are rather large (8.37 for PS and 8.44 for PST on average), but as shown on Table 4-3, tend to increase for shorter gauge length. This is however not sufficient to account for the premature failure observed experimentally, as compared to the GLS model. This highlights the limitation of this analytical approach, which was reported by other authors, since this model does not take into account the local interaction between fibers, such as when one fiber breaks, a region around this is affected. However, the model can be used as a first approach to assess the reduction in strength of the PST composite as compared to PS. As for the experimental data, the reduction is rather minor.

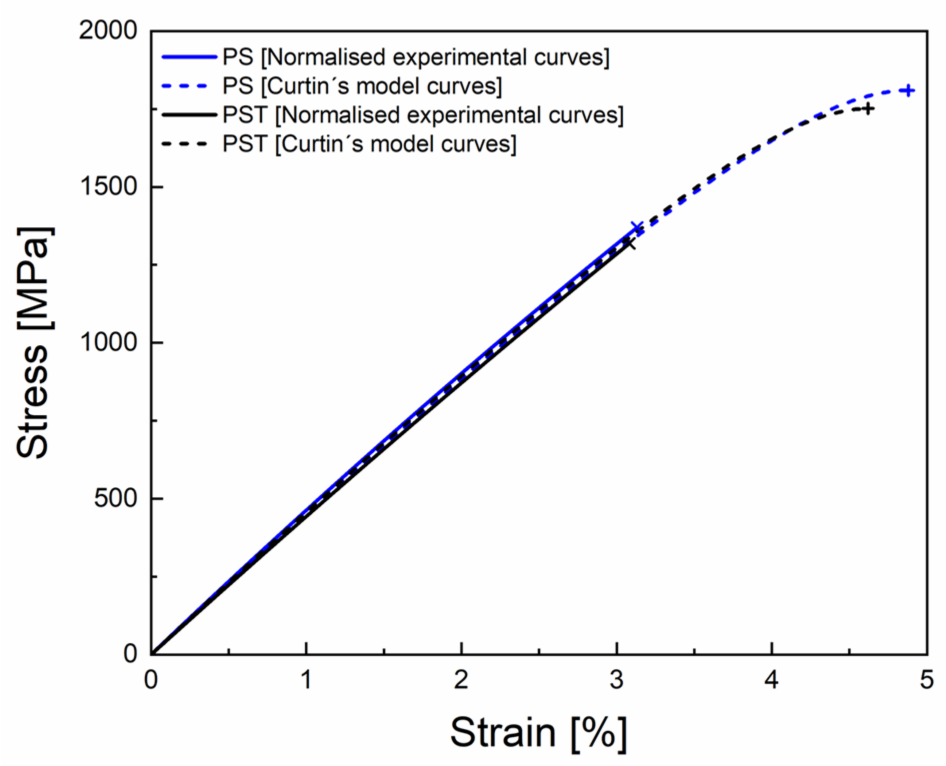


Figure 4.25 Comparison of experimental stress-strain curves and predictions from the GLS model for PS and PST composites.

4.3.5 Conclusions

This chapter evaluated the role of potential pre-treatment methods such as a mechanical pre-spreading and a thermal treatment to increase the quality of the thin-ply composites, and to increase the ability to process hybrid composites by introducing additional steps to ensure that several types of fibers can undergo a simultaneous process.

Overall, the pre-treatments led to high quality prepregs, and did not significantly compromise the fiber orientation and the handling of the prepreg. The more detailed analysis of the glass fiber prepreg demonstrated that the treatments did not affect the modulus but reduced the strength of the individual glass fibers. This translated into a slightly reduced strength of the composites, and a slightly lower achieved fiber volume fraction for the same processing conditions, but both remain within acceptable limits.

As a conclusion, for the further processing of hybrid prepregs, it was decided not to use the additional pre-treatments, and to keep to original NTPT processing and spreading method as much as possible.

Chapter 5 Tow-level thin-ply prepreg manufacturing

5.1 Introduction to tow-level hybrid thin-ply prepreg manufacturing processes

The manufacturing of thin-ply tow-level hybrid prepregs illustrated in Figure 5.1 shares several of its production stages with the manufacturing of single-fiber non-hybrid which was discussed in Chapter 4. However, it was only made possible after the development of new equipment and the adjustment of the production processing parameters. The objective was to arrange the different fiber tows side by side and guide them into the main spreading stage of the North Thin Ply Technology prepregger while maintaining the integrity of this defined hybrid pattern and avoiding disruptions or interference with subsequent production stages.

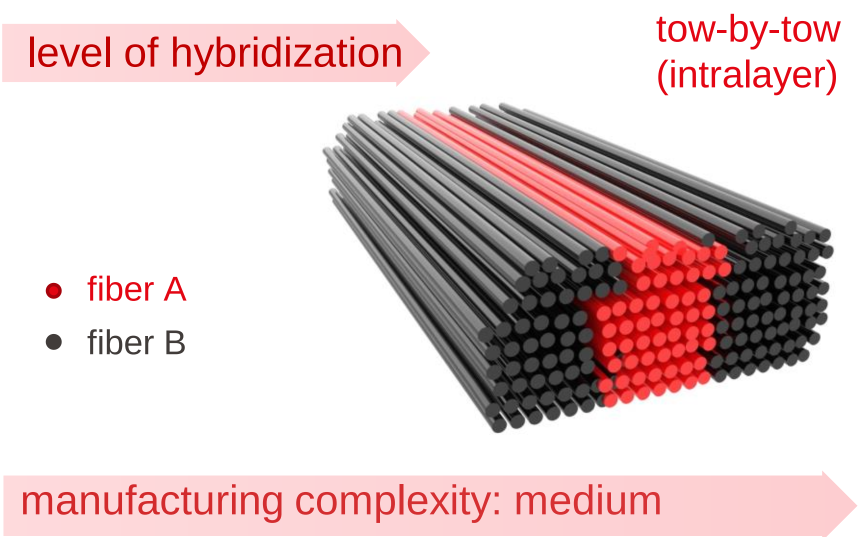


Figure 5.1 Ideal tow-level hybridization.

The critical process parameters of fiber tension, and production speed, that were involved in the production of hybrid prepregs were carefully monitored to assess their effect on prepreg uniformity, level of fiber dispersion, and the tow-by-tow hybrid pattern. The precise adjustment of these parameters allowed for improved control and resulted in tow-by-tow hybrid quality equal to that of single-fiber non-hybrid prepregs. Furthermore, this enabled experimentation with different hybridization scenarios and the successful manufacture of a wide range of hybrid prepregs with varying properties, expanding the product design space. The following sections will outline the features of custom-made equipment and modification to existing production methods that enabled the efficient and controlled manufacturing of tow-by-tow hybrid prepregs.

5.2 Fiber unwinding control system

The initial stage in the production of the tow-level hybrid prepreg involves configuring the fiber feeding stage of the production line by arranging the bobbins of the two different fibers on a creel stand. The experimental prepreg line of North Thin Ply Technology in Renens, Switzerland, has a creel stand with 12 available bobbin holders as shown in Figure 5.2, which determines the maximum width of the produced prepreg, as well as the tow-by-tow hybridization patterns and fiber ratios that can be simultaneously spread in a single prepreg tape. Meanwhile, the full-scale production line at the plant in Zory, Poland provided the necessary capabilities for the fabrication of all tow-by-tow hybrid types required for the present study.

At this production stage, two critical parameters, the fiber tension, and the production rate/speed of production, can be controlled. Unlike the production of non-hybrid prepregs, where these parameters remain constant throughout the production and can be preset with simpler devices, such as the custom-made system of pneumatic actuators that act as breaks on the creels Figure 5.3a. the manufacturing of tow-by-tow hybrid prepregs often required more frequent, precise, and independent online control of these parameters in order to stabilize the production process.

The independent controllability of the production parameters is imposed by the simultaneous handling of two dissimilar fibers and ensures that each fiber type is processed optimally and always gives the right tension as well as the required processing time (spreading and impregnation duration).

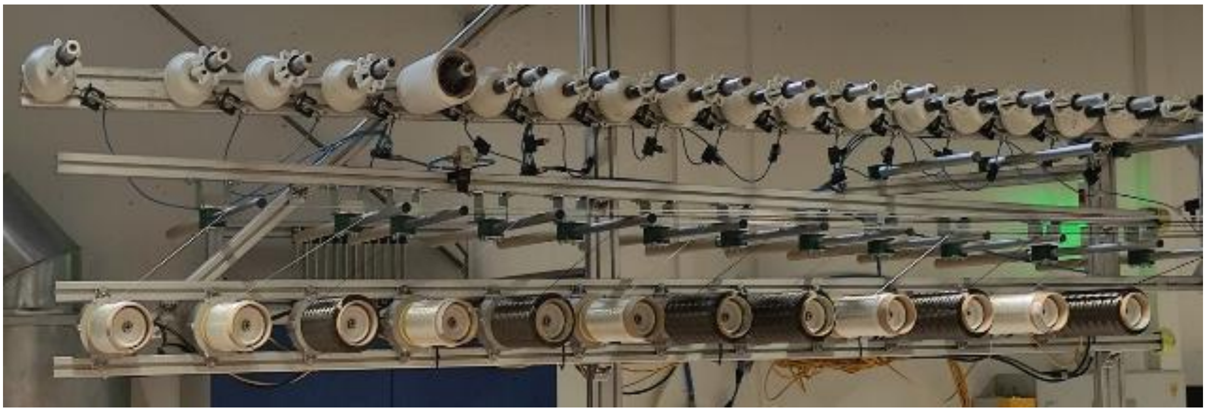
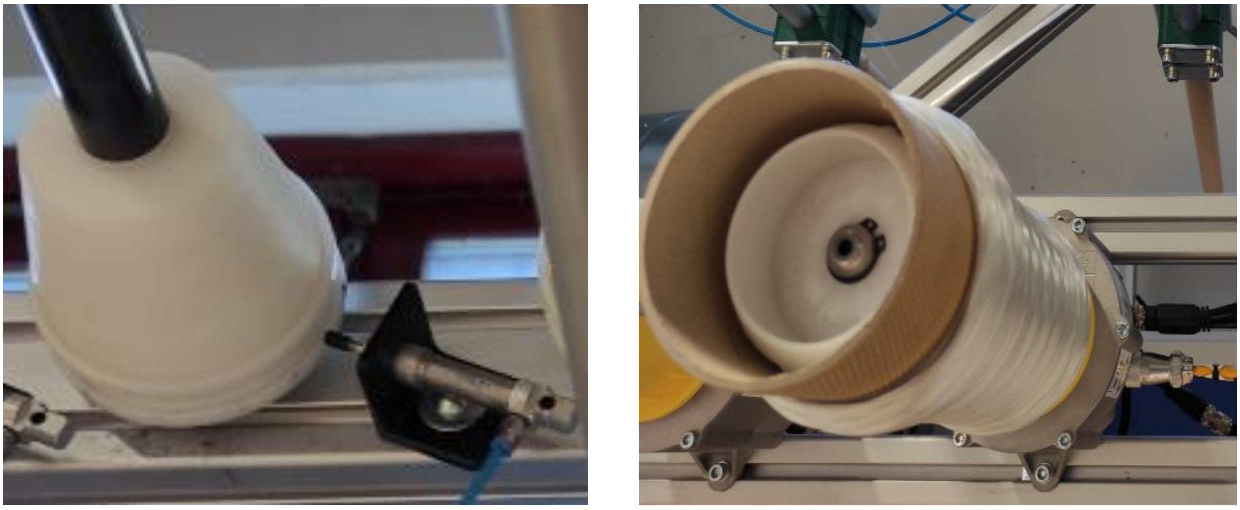


Figure 5.2 Photograph of the experimental setup to illustrate the fiber creels of a creel stand and show two different types of unwinding units.

This independent controllability could only be achieved by using an electronically controlled unwinding unit (EGA) Figure 5.3b. EGAs are adopted from the textiles industry and are exclusively intended to unwind spools or bobbins of thread, thread-type fibers, yarns, ply yarns, and technical textiles, such as carbon and glass filaments, with a defined yarn tension and speed. Any variation in production can be detected immediately by sensors within the unit and promptly compensated for, as an EGA unit functions both as a brake and driving engine. The use of EGA unwinding units enhances the control of the prepreg line configuration as they can be divided into multiple individual groups, each with its unique recipe (set of tension and unwinding speed parameters). By adjusting/alternating the tow tensions between the different fiber types, the flexibility of the spreading method increases, resulting in higher fiber dispersion or stabilization of the required tow-by-tow pattern.



a b

Figure 5.3 a) custom-made pneumatic unwinding unit, b) electronically controlled unwinding unit (EGA).

5.3 Fiber combs

The key components that facilitated the production of tow-level hybrid thin-ply prepregs were the fiber combs Figure 5.4. These fiber combs were positioned between the fiber feeding stage and the main spreading stage and served as a fiber guiding device for the fibers that were unwound from the creels into the prepregger. These custom-made pieces of equipment were specially designed to fulfill two primary functions. Firstly, to ensure the proper spatial arrangement of the tow dissimilar fibers, and secondly, to determine key characteristics of the final product, such as the fiber areal weight, the width of the individual tows, and the total width of thin-ply prepreg. The fiber combs also helped to impose a tow-by-tow pattern Figure 5.5, Figure 5.6 and guided the dissimilar fibers that were arranged side by side into the main spreading stages of NTPT Figure 5.7, which is considered a “black box”. There, the dissimilar fibers were spread simultaneously, and the energy offered by the spreading mechanism forced the tows to spread until they met their neighboring tows, resulting in a well-defined tow-by-tow hybrid pattern. This pattern was stabilized on the backing paper during the impregnation stage that followed, creating a single layer of tow-by-tow hybrid thin-ply prepreg.

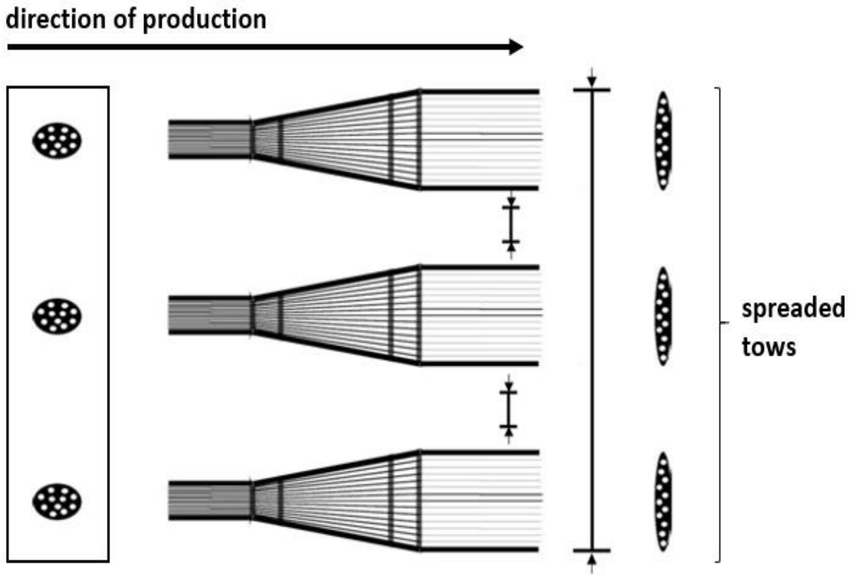


Figure 5.4 Schematic of a fiber comb.

The design of the fiber combs can take various forms, such as a metal steel plate with drilled guiding openings or can consist of a frame with guiding rollers or metal bars. The metal surfaces of the fiber combs that come in contact with the fibers should be polished to reduce friction. Rubber O-rings are often installed at the guiding holes that are opened on the metal to prevent potential damage to the fiber passing through it Figure 5.6. In conclusion, fiber combs have been proven to be essential elements in the hybrid prepreg production process. They are highly adaptable, low-cost components that allow for quick adjustments to the fiber arrangement. In combination with the precise adjustment of other manufacturing stages in the prepreg production process, they greatly contribute to the efficient and controlled fabrication of high-quality tow-by-tow hybrid thin-ply prepregs Figure 5.8. When designing the fiber combs, factors such as the materials used, the size and shape of the guiding openings or rollers, and the smoothing of the metal surfaces in contact with the fibers should be considered.

Figure 5.5 Schematic of simultaneous spreading of two dissimilar fibers in a tow-by-tow hybrid configuration.

Figure 5.6 Custom-made comb used for simultaneous fiber spreading (tow-by-tow hybrid spreading) and hybrid thin prepreg manufacturing.

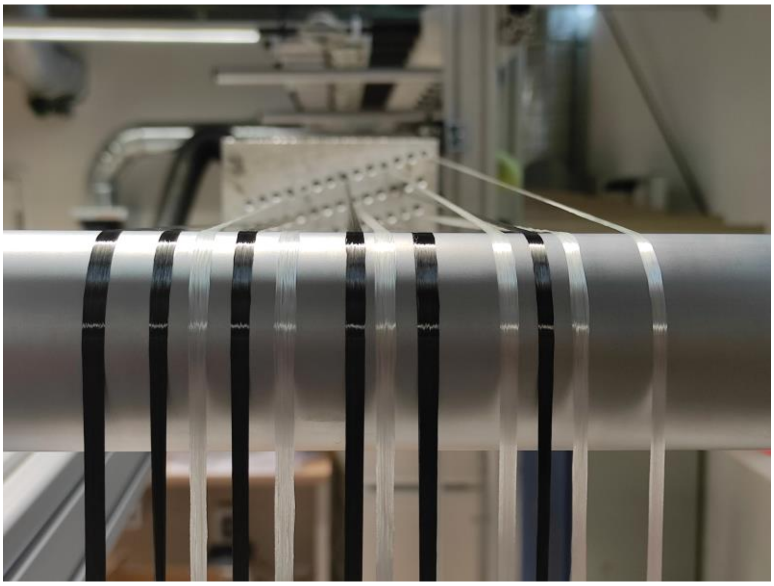


Figure 5.7 Tow of carbon and glass fibers arranged side by side before entering the main tow spreading of the prepreg process.

Figure 5.8 Tow-by-tow glass-carbon hybrid prepreg at the final stage of the production line.

5.4 Manufacturing of tow-by-tow hybrid prepregs

Three types of tow-by-tow hybrid prepregs were manufactured for this study with the simultaneous fiber spreading of dissimilar carbon fibers as presented in Figure 5.9, Figure 5.10, Figure 5.11. The quality is visually acceptable, and it is not possible to distinguish the two types of fibers.

Figure 5.9 Carbon (HR40) – carbon (34-700) tow-level thin-ply hybrids tow-by tow Test 1: ${}^{31}T^{90}$.

Figure 5.10 Carbon (HR40) – carbon (34-700) tow-level thin-ply hybrids tow-by-tow Test 2:$^{25}T^{90}$.

Figure 5.11 Carbon (HR40) – carbon (34-700) tow-level thin-ply hybrids tow-by-tow Test 3:$^{31}T^{45}$.

5.5 Manufacturing of hybrid tow-by-tow laminates

The optimization of fiber spreading and impregnation techniques enabled the production of high-quality hybrid prepregs at tow-by-tow configurations and several hybrid patterns. The level of controllability gained over the manufacturing process is illustrated in Figure 5.12. The parameters that can be controlled are the fiber type, tow count and final thickness T of the tape. These are linked, and for a given sought thickness T, the tow spreading will be dictated by the initial tow size. As a result, the width of each spread tow Wc and Wg is directly related to the thickness T, and can thus be quite large for usual tow sizes (12 and 24k). Following the composite manufacturing methods that were established during the manufacturing optimization phase of the project, different hybrid composite plates were produced and thoroughly examined to reveal the characteristics of this new type of material as presented in Figure 5.13. Of particular interest were areas in the borders of dissimilar fibers where defects were more likely to appear. Finally, the microstructural analysis of the cross-sectional area of different hybrid tow-by-tow composites reveals well-aligned fibers, limited void content, and good fiber dispersion.

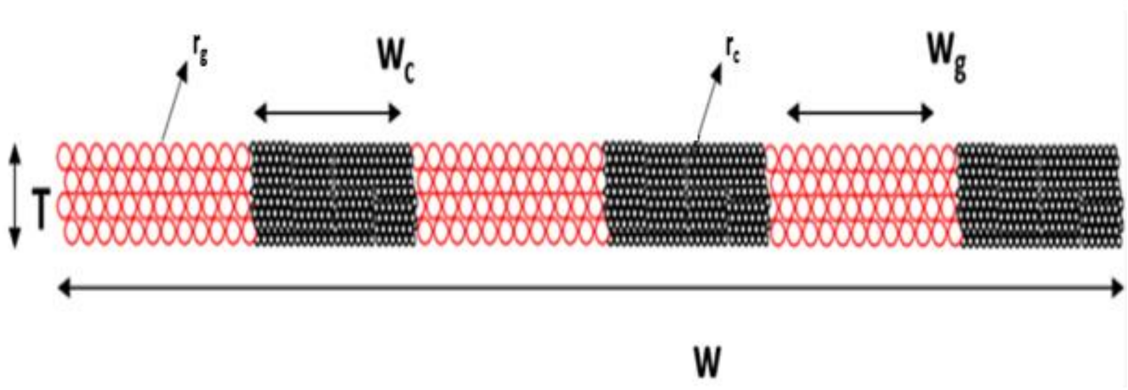


Figure 5.12 Level of controllability gained over the tow-by-tow thin-ply prepreg production process.

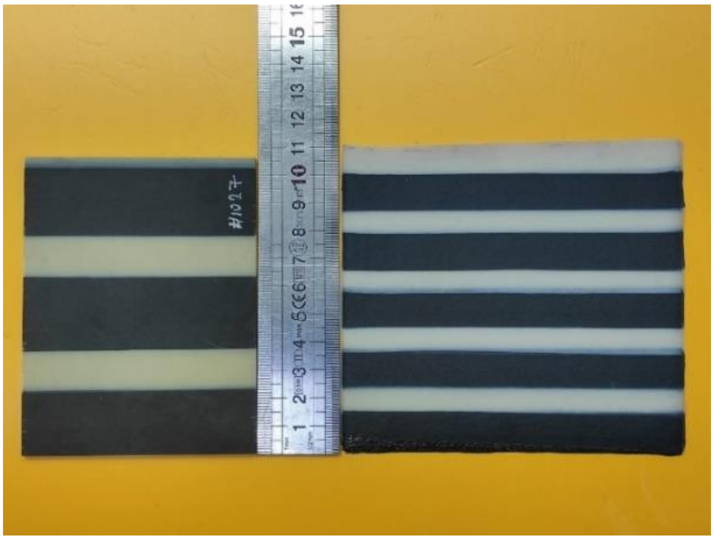


Figure 5.13 HS40 40% - E-glass (60%) 54 gsm tow-by-tow hybrid. E-glass tex 1200 (left) Elgass tex 600 (right). Microstructural features of tow level hybrids.

5.6 Conclusions on the tow-by-tow manufacturing process

The development and adaptation of a custom-made comb system to separate the tows and align them for further spreading and impregnation was successful. Several types of prepregs were produced, with glass and carbon, or with two carbon grades, that will be characterized in Chapter 7.

Chapter 6 Fiber-level hybrid prepreg manufacturing

6.1 Introduction to fiber-level hybrid thin-ply prepreg manufacturing processes

Several different approaches were tested for the fabrication of fully co-mingled (fiber-level) hybrid thin-ply prepregs as illustrated in Figure 6.1. It was anticipated from the beginning of the study that these approaches would involve a significant increase in the manufacturing complexity, to get full controllability and consistent results. North Thin Ply Technology employs a proprietary fiber spreading and impregnation method that had never been previously tested for its potential to produce fiber-by-fiber hybrid prepregs. The initial fiber-by-fiber hybridization attempts were performed on the experimental prepreg line of the North Thin Ply Technology in Renens, Switzerland through a series of manufacturing experimentations. The objective was to explore the capabilities and limitations of the existing thin-ply prepreg production line and investigate the possibility of utilizing the available processes and equipment. The overall goal was that any successful processing method could then be easily implemented into large-scale industrial production, after the necessary adjustments with minimal requirements for new processing components. This would result in significant benefits in terms of efficiency and cost reduction.

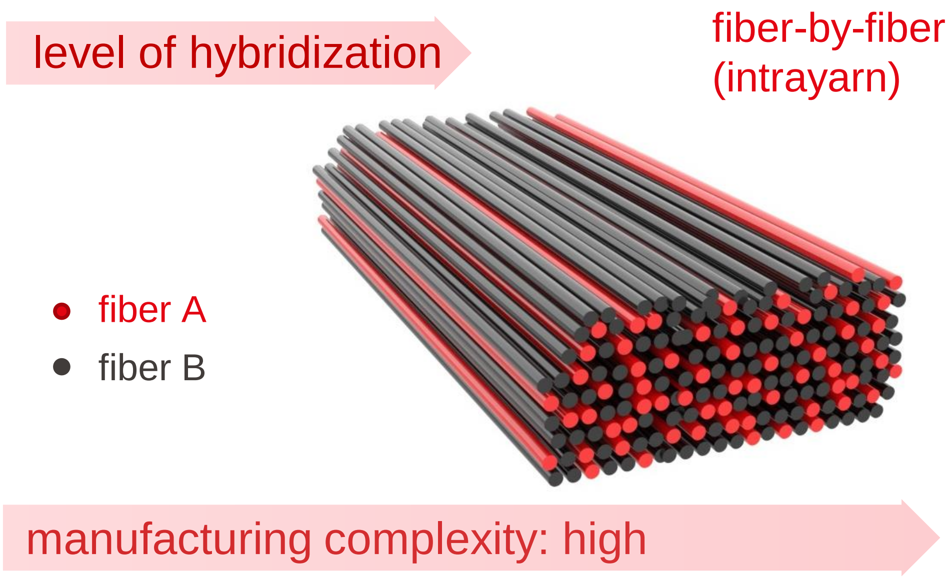


Figure 6.1 Ideal fiber-level hybridization.

Despite extensive efforts to optimize the existing prepreg manufacturing processes, including those performed as part of earlier stages of this research, the goal of achieving fiber-by-fiber hybridization proved challenging and beyond the capacity of the existing production method of North Thin Ply Technology. This chapter will discuss the attempts of simultaneous fiber spreading for the fabrication of fiber-level hybrid prepregs and the manufacturing challenges that were encountered. Then all production modifications and adjustments that were made before concluding that this method could not bring the desired level of fiber hybridization and attempting a different approach will be presented. The chapter will also introduce a novel hybrid thin-ply manufacturing method, the calendering process, which was proposed as an alternative for achieving the desired level of fiber hybridization. A comprehensive analysis will be given on how the new method differs from the simultaneous spreading of dissimilar fibers, and all the optimization processes, from the conceptual stage up to the implementation to the industrial production of NTPT, will be discussed in detail. Finally, the chapter will present the results of the hybridization achieved through the newly developed calendering process and compare them to the results obtained through simultaneous fiber spreading. The limitations of both processes for industrial manufacturing of fiber-by-fiber thin-ply prepregs will be discussed and suggestions for future work will be provided.

6.2 Fiber-level hybridization with simultaneous fiber spreading methods

Typical, tow/fiber spreading methods for the production of thin-ply prepreg materials have been primarily designed to handle one type of fiber at a time. There are some exceptions reported in the academic literature as shown in Figure 6.3, however, these methods are limited in terms of volume of production, only achievable through laboratory-scale experiments, and their results are often unpredictable with little to no control over the final prepreg’s characteristics, such as fiber volume fraction and resin content. Their focus is to achieve any possible level of fiber mixing with little to no controllability over the microstructural profile of the produced prepregs and later the hybrid composite laminates. As for the industrial prepreg manufacturing methods, no process or commercial product has been reported to combine thin-ply and fiber-level hybrid characteristics in one single ply of prepreg material.

Exploring the use of existing industrial fiber spreading methods to achieve mingling at the fiber level in prepreg production may be viewed as unconventional. Most of these methods, such as those utilized by North Thin Ply Technology (NTPT), have been primarily designed to break down larger tows into smaller bundles and align fibers, rather than mixing them. Therefore, spreading and simultaneously mixing fibers pose significant challenges due to process design and/or machine (prepregger) limitations and conflicts. Additionally, the experimentation with hybrid tows of fibers that could potentially be compatible with the NTPT process for manufacturing fiber-level hybrid prepreg was beyond the scope of this research, due to the high cost and limitations imposed by the predefined fiber types and fiber ratios of commercial hybrid tows.

6.3 Testing and optimization of simultaneous fiber spreading prepreg manufacturing methods

This work on the simultaneous spreading of dissimilar fibers focuses on the handling and processing of the fibers before they enter the proprietary main fiber spreading and impregnation stages of NTPT, which are seen as "black boxes". All the experiments were based on alternating the configuration of the production parameters before the main fiber spreading stage, such as the tow feeding stage and tow creel arrangement, the speed of production, and the use of fiber pre-spreading stages that were found to be efficient at previous stages of the study to fine-tune the process. However, there was not any set of production parameters that could bring the targeted level of fiber mixing and microstructural controllability. The following sections will briefly present some of the optimization attempts that were performed on the experimental prepreg line of NTPT.

6.3.1 Independent control of fiber tension for the different fiber types

This experimental set-up aimed to effectively arrange two distinct fiber tows on top of each other by having them pass through the same narrow openings of the custom-made guide combs as shown in Figure 6.2. The objective was to stabilize this system of tows in a "sandwich tow" configuration and guide it into the main spreading stage without splitting and extensive fiber misalignment, hoping that the field of forces applied there will result in some level of interpenetration and fiber co-mingling. Close attention was paid to the monitoring and precise control of the speed of the production and the tow applied on the tow of fibers. These two production parameters were found to have particular importance for ensuring a uniform flow of the "sandwich of tows" and the efficient operation of the main spreading stage. The use of electronically controlled unwinding units (EGA), the operation of which was described in detail in Chapter 5, enabled the experimentation with a wide range of production parameters (T: 0-1500N, S:1-15 m/min) but most importantly enable to differentiation of the tension that was applied on different types of fibers.

The approach of simultaneous fiber spreading targeting a fiber-level hybrid composite configuration was primarily inspired by the work of Diao et al. [129], who reported that some degree of fiber comingling could be achieved when two different types of fiber tows (glass and carbon) were exposed simultaneously under the same field of forces applied by a pneumatic fiber spreader. However, despite several iterations and different production setups, this concept could not be successfully replicated with the materials and equipment used in this study.

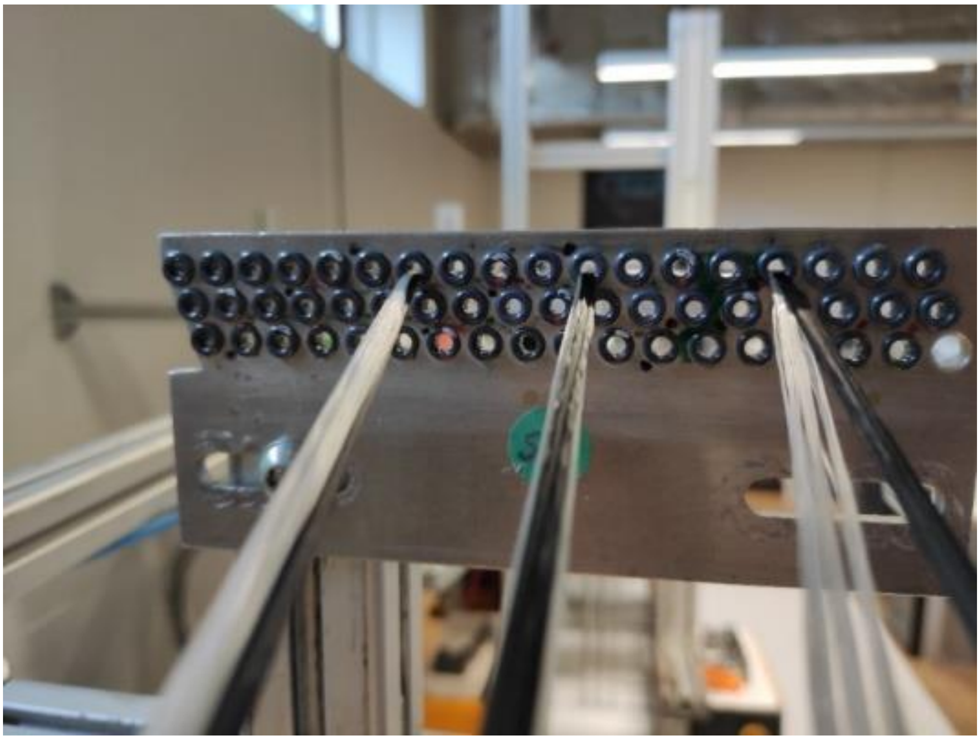


Figure 6.2 Simultaneous spreading targeting to fiber-level hybridization. Simultaneous pre-spreading of dissimilar tows.

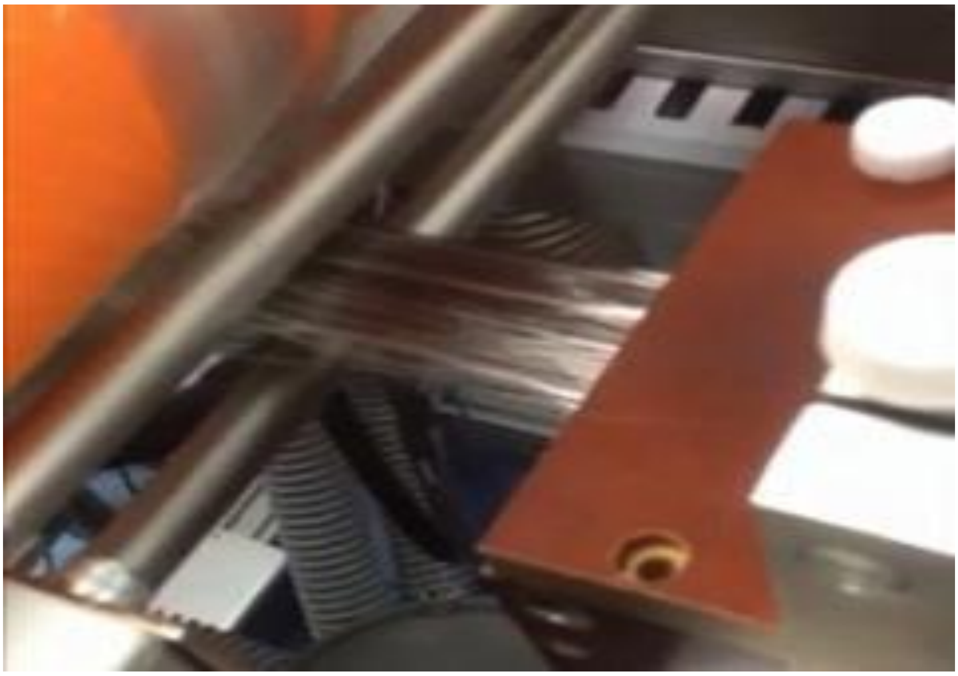


Figure 6.3 Simultaneous fiber spreading apparatus. Narrow tapes of carbon-glass hybrid prepregs manufactured with the simultaneous fiber spreading method.

6.3.2 Fiber pre-spreading of the different fiber types

This experimental spreading process aimed to assess the potential of the fiber pre-spreading stage that was developed at an earlier phase of this study for the production of thin-ply prepregs with a fiber-level hybridization. The process involved pre-spreading each type of fiber individually before bringing them together and subjecting them to the main fiber spreading stage of the North Thin Ply Technology process for further mixing. The fibers were unwound from creels and divided into two groups of the same fiber type using a "splitter comb." Each group was then guided to a pre-spreading stage where the configuration could be adjusted if needed. Once a sufficient level of pre-spreading was achieved (50-100%), the groups of fibers were combined to create a system of dissimilar pre-spread tows in a "sandwich" configuration and then guided into the main spreading unit of the prepreg. The process is illustrated in Figure 6.4.

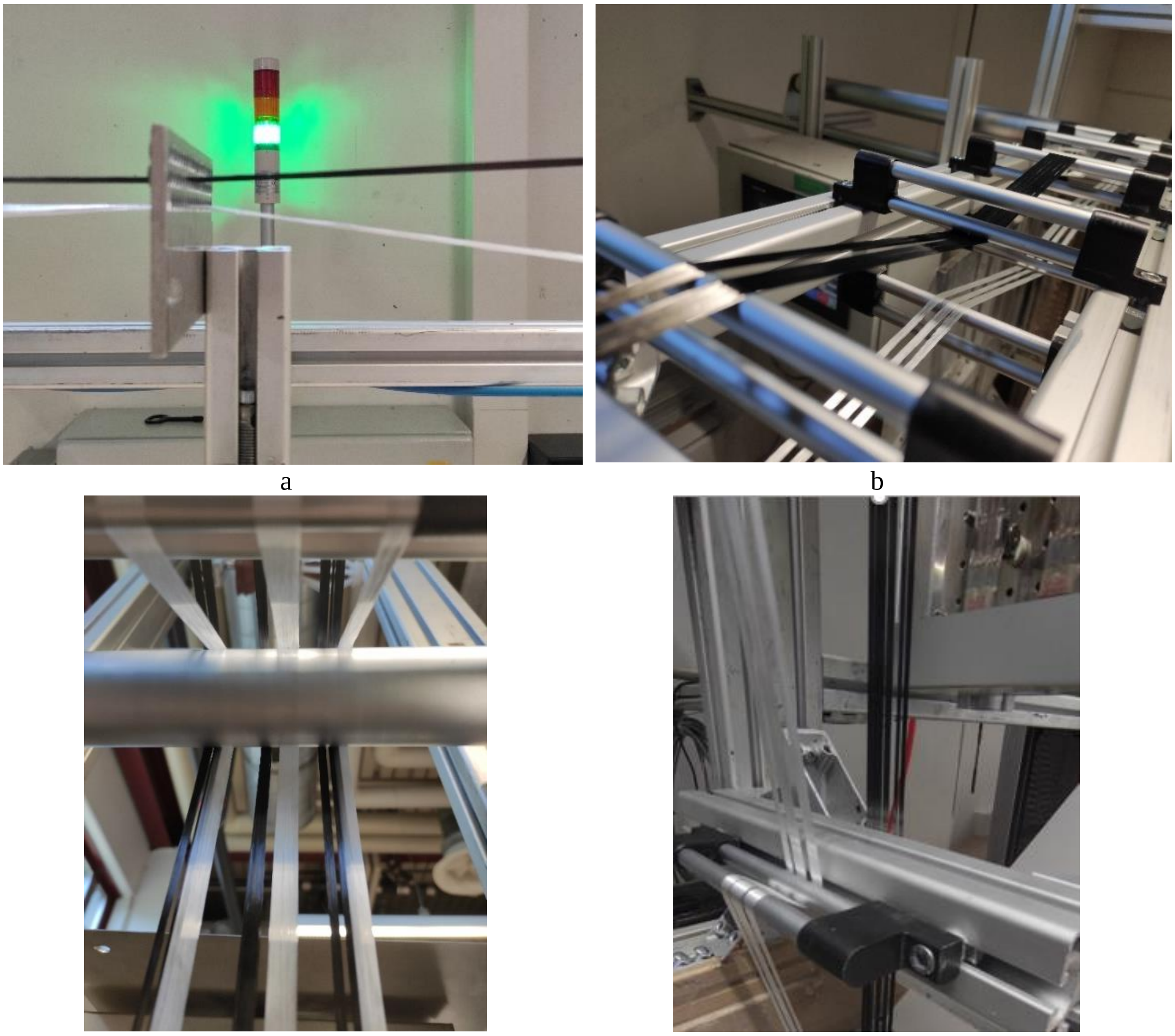


Figure 6.4 Pre-spreading setups at different stages of the production line. a) splitter comb. b) pre-spreading of two different types of tows. c) bringing the two pre-spread tows of fibers together d) guide the tow tows into the main spreading zone for further fiber spreading.

6.4 Results of the hybridization attempt with the simultaneous fiber spreading method

The goal of this part of the research was to explore the potential of the existing fiber spreading methods of North Thin Ply Technology, including all the newly custom-made equipment that was developed for this work for the manufacturing of uniform hybrid thin-ply prepregs with fiber-level mixing. However, despite numerous attempts to improve the existing NTPT production processes through established techniques and the combination of the most promising methods, consistent and satisfactory results were not achieved. These approaches were found to be too complex and required extended pre-spreading stages to be added to the production line before guiding the arrays of fibers into the main spreading unit of the prepregger. One of the significant disadvantages of these methods was the need for regular and precise fine-tuning of the process parameters during production, which increased the risk of quality inconsistencies dramatically. However, the effort put into optimizing existing manufacturing methods provided valuable insights into the fiber spreading mechanisms (limitations) and important feedback for the future development of hybrid fiber spreading processes. The goal was to attain a uniform system of pre-spread fibers. The following sections will briefly present the fiber hybridization results.

6.4.1 Results of simultaneous fiber spreading with tension control

Attempt for simultaneous spreading of E-glass and carbon fibers with tension control are presented in Figure 6.5. Only a small level of hybridization could be achieved with this method. The co-mingling is uneven through the width of the prepreg and it was not possible to obtain a usable prepreg with this method.

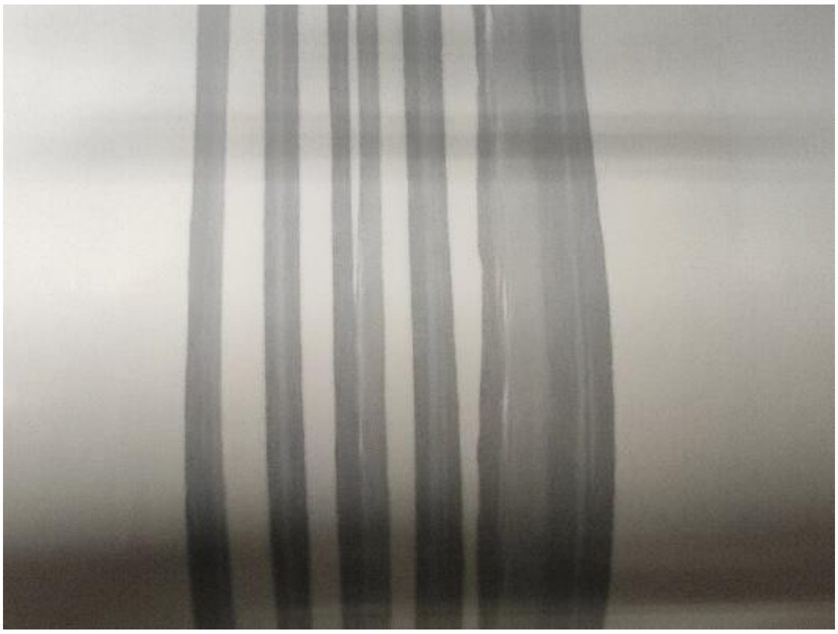


Figure 6.5 Uneven fiber co-mingling of E-glass and carbon though the width of the thin-ply tape making the resulting material unusable. Result of the simultaneous fiber pre-spreading and tension control.

Several experimentations were conducted with multiple sets of production parameters, including the configuration of the pre-spreading stage, the number of bars, the pre-spreading path, the length of the pre-processing stage, and its positioning relative to the entrance of the fiber pre-spreading zone. At the same time, the fiber tension was precisely controlled to ensure uniform alignment and consistent quality of the final product. The final product was more spread and mingled compared to the simple version tried before. However, none of these adjustments helped to optimize the process to achieve the desired level of fiber hybridization in the final prepreg material as can be visually observed in Figure 6.6 and Figure 6.7.

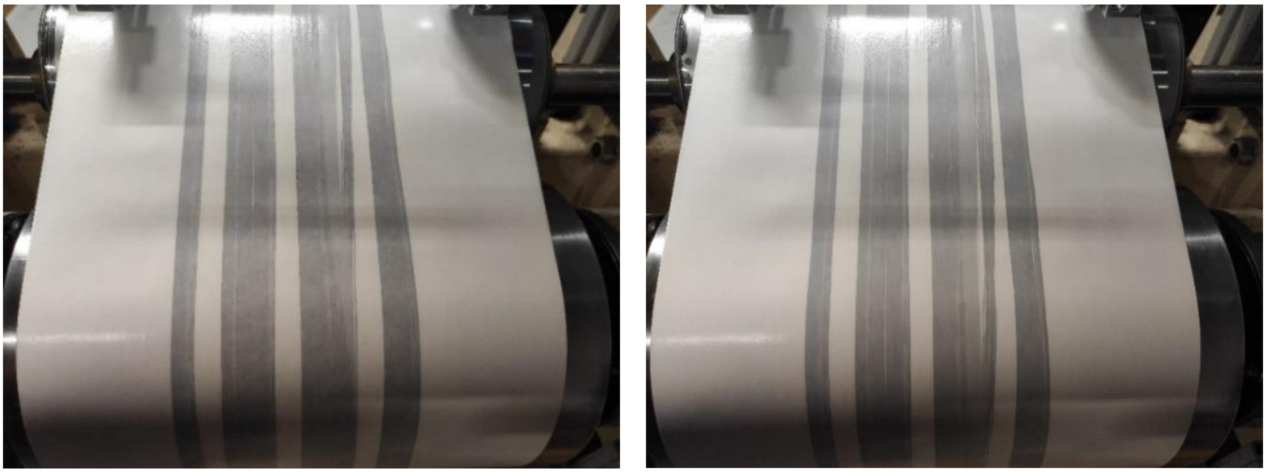


Figure 6.6 Attempt for simultaneous spreading of E-glass and carbon fibers with fiber pre-spreading and tension control. The pre-spreading increases the comingling at random areas through the width of the prepreg but still the level of mixing is not satisfactory.

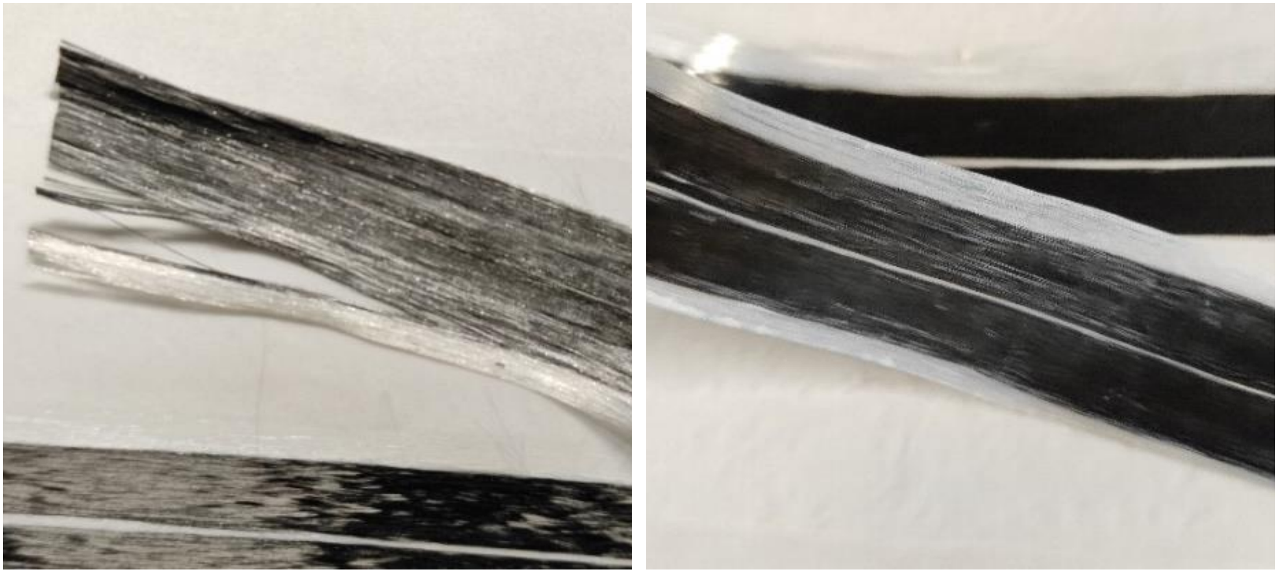


Figure 6.7 Thin strip of E-glass-carbon hybrid prepreg manufactured with fiber pre-spreading and fiber tension control. It can be observed that the level of mixing is not consistent and the quality of the prepreg does not allow further processing.

6.5 Calendering process for the manufacturing of fiber-level hybrid prepregs

A novel thin-ply prepreg processing method, the calendering process, was proposed and tested as an alternative solution to overcome the manufacturing challenges faced in the simultaneous fiber-spreading processes. Calendering is a mechanical shaping process commonly used to produce flat, continuous thin sheets of material. In this process, the materials are passed through a series of counter-rotating rolls at high temperatures and pressures, which flatten them and impart the desired characteristics. The uninterrupted operation of a calendering machine is ensured by accurately adjusting various process parameters such as distance, temperature, and rotation speed of the rolls. The calendering processes have been widely used in various industries, including the textiles industry as a finishing technique, due to its ability to give specific characteristics to the fabrics by changing the structure of the fiber yarns. In this work, a novel composite manufacturing method was inspired by the concept of calendering processes and adapted to work with thin-ply prepregs to create hybrid composite architectures with hybridization down to the fiber level. During the development of this new manufacturing method, the effects of the calendering processing parameters on the characteristics of the final product were determined through microstructural analysis and mechanical testing of the hybrid composites produced with calendered thin-ply hybrid prepregs.

6.6 Development of a laboratory-scale calendering apparatus

To explore the potential of the calendering process for the manufacturing of fiber-level hybrid thin prepregs and composite materials, a small-scale custom-made experimental calendering apparatus was initially set up at NTPT Renens, Switzerland for the proof of concept. The main components of this apparatus were two steel counter-rotating calender rollers. The apparatus was designed to allow precise adjustments of key production parameters such as the temperature of the rollers, the gap between them controlling the pressure, and the production speed Figure 6.8.

The calendering process was implemented by guiding two different single-fiber thin-ply prepregs, used as intermediate prepreg, between the tight gap of the calender rolls at elevated temperatures and pressures. This forced the two types of fibers to be flattened and mingled together, resulting in fiber-level hybrid architectures. The intermediate single-fiber prepregs used as intermediate materials for the calendering process were manufactured with the processes described in detail in Chapter 4, "Non-hybrid thin-ply prepregs," and have predefined characteristics such as fiber areal weight, ply thickness, and resin content. These intermediate single-fiber rolls can be of different fiber types, such as carbon-glass (e.g., E-glass/HS40), which were used at the initial stages of the process during the development of the calendering process. Alternatively, they can be of the same fiber type, but of different grades, such as carbon-carbon (e.g., 34-700 and HR40), which were used for the main part of the development and study of hybrid thin-ply prepregs and composite materials in this work.

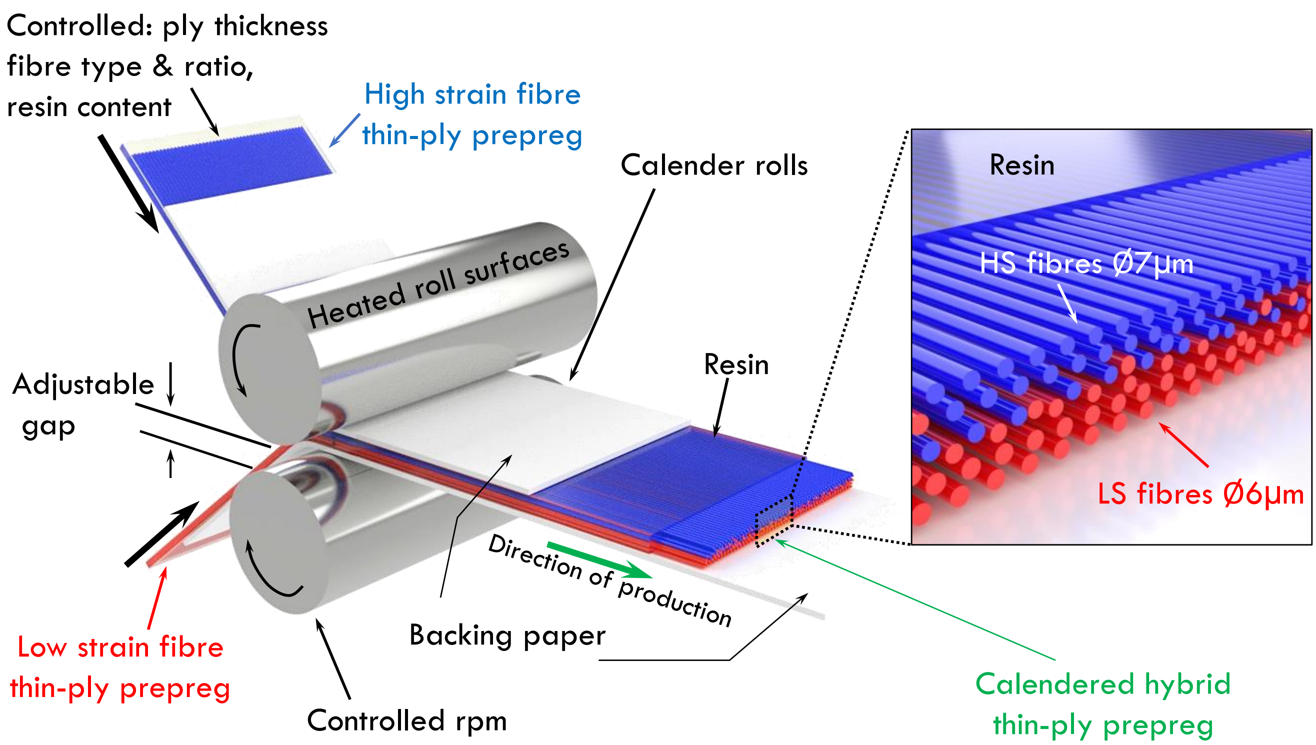


Figure 6.8 Calendering experimental apparatus for fiber-by-fiber thin-ply prepreg manufacturing.

6.7 Industrial scale calendering process

Following the successful proof of concept and preliminary quality control of the new calendered materials, the next objective was to transition from a small laboratory-scale calendering apparatus to a larger-scale calender and to industrialize the process, enabling the production of larger quantities of hybrid thin-ply prepregs in a time- and cost-efficient manner while achieving equivalent results in terms of fiber-level hybridization. To achieve this goal, an industrial calendering apparatus was set up at the production plant of NTPT in Zory, Poland, based on the experience acquired during the development phase of the experimental small-scale calender in Renens, Switzerland. The industrial calendering apparatus had the significant advantage of being able to process larger quantities of materials, with the ability to process materials up to 300 mm wide and approx. 150 m long.

6.7.1 Calendering processing parameters

The calendering process was performed with a set of larger steel calender rolls Figure 6.9 with different processing parameters to manufacture the hybrid thin-ply prepregs for this work. More specifically the processing temperature, controlled with heating elements in the core of the rolls, was set to 80°C which was found as an optimal temperature for lowering the viscosity of the thermosetting TP 415 resin and allowing some fiber movement that could promote the fiber mingling and hybridization while still being well below the curing temperature of the resin.

Of particular importance for the present study is the pressure setting of the calendering process which adjusted with a set of pneumatic pistons acting on the axis of the calender rolls. For this study, the calendering process was performed with two different production scenarios settings of the pressure of production parameters, namely high-pressure (HP) rollers and low-pressure (LP), the exact pressure values however cannot be reported due to confidentiality limitations. Finally, the speed of the calendering production was set to 4m/min which allowed the pressure of the rolls to be maintained consistently through the production process, which was important for ensuring the quality and consistency of the final product processed with both high (HP) and low (LP) pressure settings.

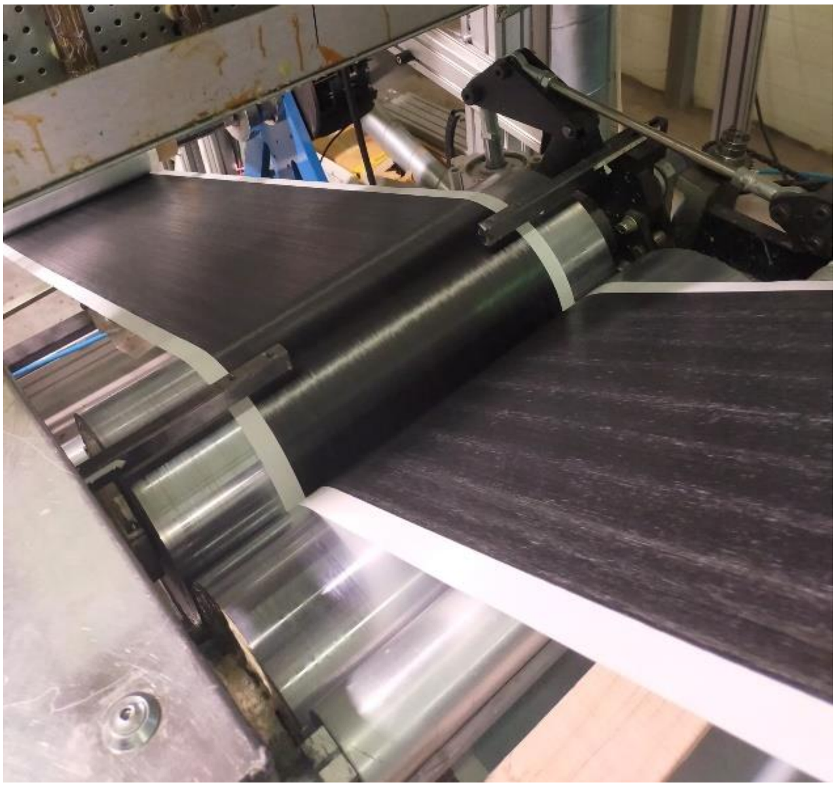


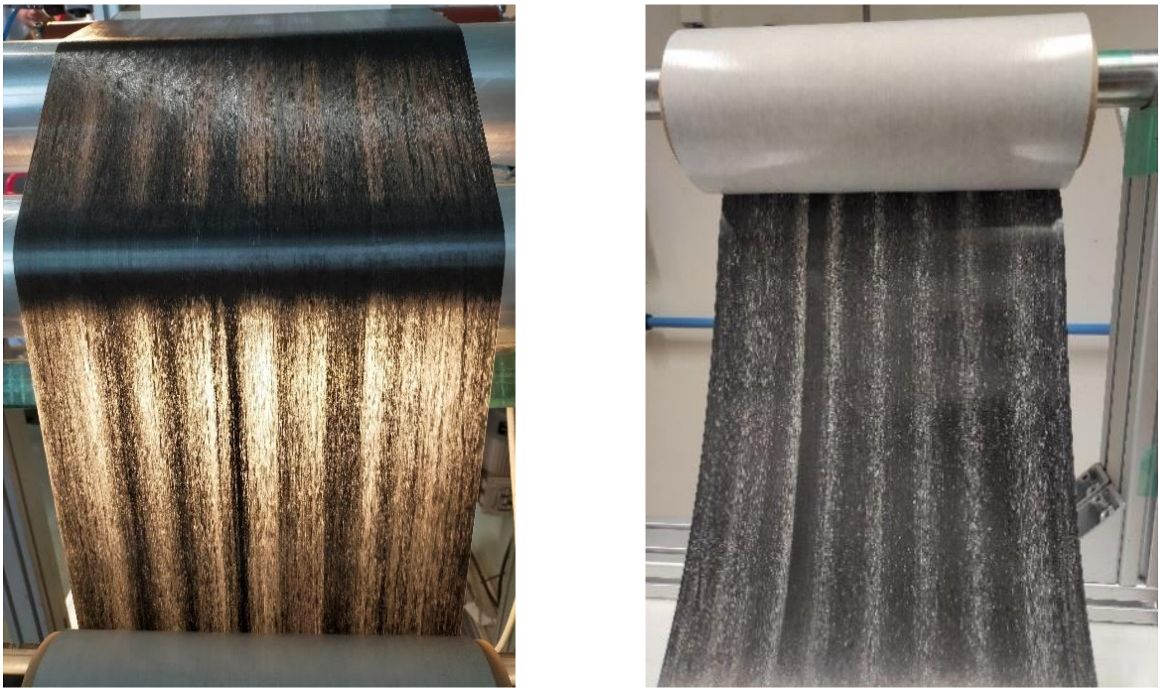
Figure 6.9 Industrial scale calendering set-up at NTPT facilities in Zory, Poland.

6.8 Comparison of the two different fiber-level hybrid prepreg manufacturing approaches

An important aspect discussed in this work is the trade-off between the advantages and disadvantages of using the calendering process for manufacturing hybrid thin-ply prepregs and composite materials. Initially, the calendering method was thought to be time-consuming due to the need for additional manufacturing steps and longer production cycles. The different intermediate thin prepreg materials had to be manufactured independently, each with its specific characteristics, before being successfully calendered together to create the new hybrid prepreg. However, it was later realized that despite the need for additional equipment and processing steps, the production process parameters (temperature, pressure, production speed) were straightforward to control and easier to stabilize during production through the calender rolls. With proper adjustments, the calendering apparatus could produce high-quality hybrid prepreg tapes in large volumes.

A major advantage of this research project for the development of the calendering process was the direct access to the entire range of NTPT thin-ply prepregs and the freedom to experiment with different prepreg characteristics, such as fiber volume fraction, thicknesses, and resin content. Another important advantage was the access to ultra-thin ply prepregs with a FAW of 15 gsm, which is equivalent to three times the fiber diameter of HR40 fiber. Spreading the fibers at this level and stabilizing them with resin into a uniform thin prepreg at a low thickness pushes the manufacturing techniques to reach their limits. Spreading the fiber at such an extreme level led to a prepreg with a variable fiber density through its width (fiber-rich/poor areas), as can be observed in Figure 6.10 and Figure 6.11, with can promote the fiber co-mingling with the calendering process. These ultra-thin-ply prepregs are not commercial products yet and are mainly used by the research and development department of NTPT for the development of their product portfolio, specific custom applications (marine, automotive, aerospace), and academic research. In contrast, simultaneous fiber spreading methods, although requiring fewer processing steps and using already existing equipment or equipment that was easier to implement to the production line of NTPT, had to deal with gaining controllability over more complex or even unidentified fields of forces applied to the fibers during the fiber spreading processes. This made it difficult to produce consistent fiber-level hybrid prepreg material suitable for further testing.

In conclusion, the calendering process holds great potential for the manufacturing of hybrid thin-ply prepregs and composite materials is a promising method for the efficient manufacturing o hybrid thin-ply prepreg materials, especially at an industrial level due to the simple equipment that it requires and the straightforward controllability of the processing parameters. However, this work also aims to explore the trade-off between the advantages and disadvantages and highlight any limitations of the process (fiber damage, material degradation) by studying its effects on the thin-ply hybrid prepregs.



a b

Figure 6.10 The channels of fiber-rich areas are alternated through the width of a 300mm 15gsm HR40 thin-ply prepreg roll that was specifically produced for the development of the calendering process. a) visual inspection of the fiber distribution during production b) final product during lamination process.

Figure 6.11 Two types of carbon (left 30 gsm 34-700, right: 15 gsm HR40) thin-ply prepregs guided between the tight gap of the calendering apparatus. Areas with different fiber densities can be observed on the ultra-thin 15 gsm prepreg.

6.9 Laboratory scale calendering in the transverse direction

After the successful implementation of the calendering apparatus, at both a laboratory and industrial scale, the next step was to experiment with the optimization of the method. To further advance the calendering process, the next step was to explore the possibility of calendering in the transverse direction, as all previous calendering methods were limited to calendering in the direction of the fibers. This was since the rolls of thin prepregs were wound and guided in the calendering process in the longitudinal direction. With this new approach, the goal was to determine if calendering in the transverse direction would result in improved fiber-level hybridization and overall mechanical performance of the hybrid thin-ply prepregs as illustrated in Figure 6.12. To perform the transverse calendering process, a modification was necessary due to the lack of a proper machine for laboratory-scale transverse calendering. The solution was to modify a Wessex Resin and Adhesives Ltd. (UK) prepregger, which was then utilized as a calendering apparatus Figure 6.13. The modification involved adjustments to the machine's components and the addition of new ones to adapt it to the requirements of the transverse calendering process. This innovative solution allowed for the efficient implementation of the transverse calendering process in the laboratory setting, enabling the investigation of its potential for producing hybrid thin-ply prepregs.

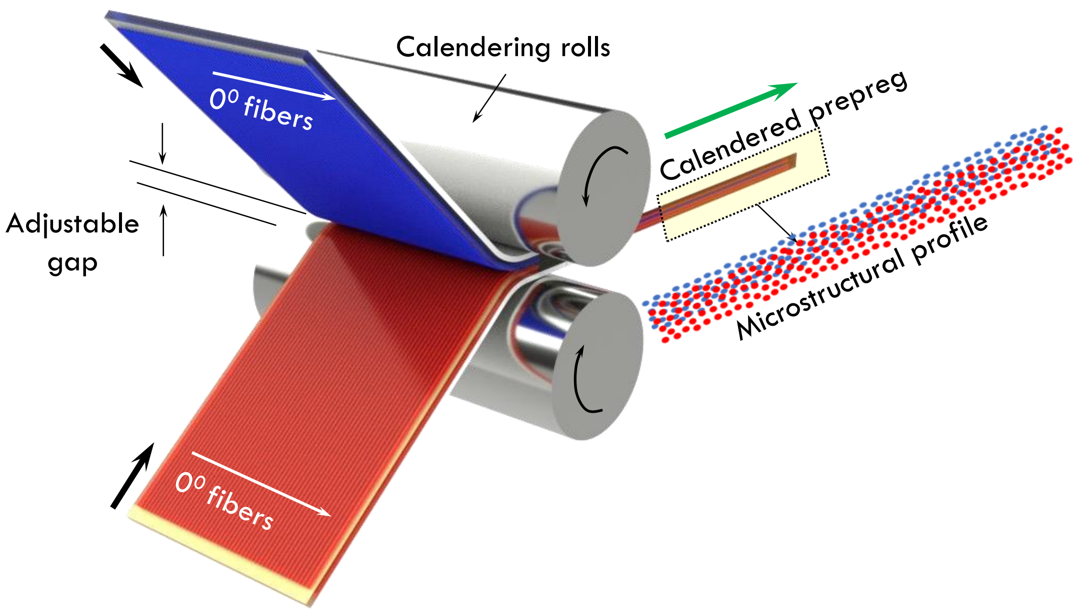


Figure 6.12 Calendering in the transverse direction of the fibers.

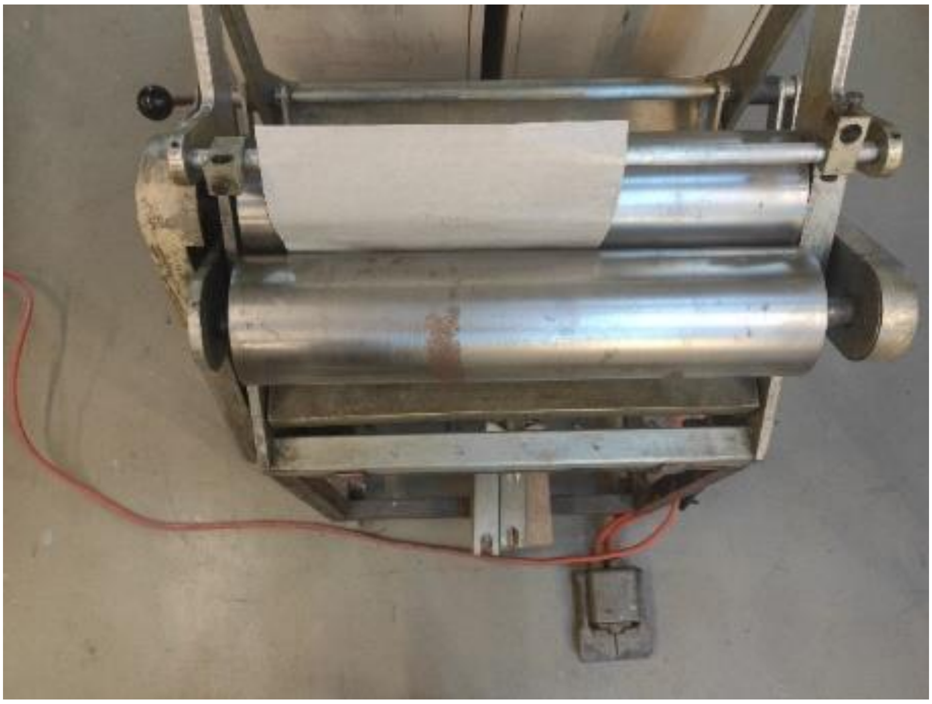


Figure 6.13 Calendering apparatus Wessex Resin and Adhesives Ltd. (Made in England) prepreger that was modified to be used as a calendering apparatus.

6.10 Conclusions of the calendering process

The concept of simultaneous fiber spreading of dissimilar fibers for achieving fiber-by-fiber hybridization within the main spreading stage of NTPT was investigated. Additionally, all the custom-made equipment and production stages that were developed for the fabrication of tow-level hybrids at previous phases of the study were also put into extensive testing to examine their potential for bringing improvements in fiber-level hybridization. The possible benefits of achieving improved production efficiency and cost reduction by utilizing existing equipment and processes made simultaneous fiber spreading approach a promising candidate for the manufacturing of fiber-level hybrid prepregs. However, the manufacturing challenges encountered in the process, the inconsistency of the quality results and the limited level of fiber mixing, led to further investigation for methods that could fit for the purpose of this study. A novel prepreg processing method, the calendering process was proposed as an alternative method. Throughout a comprehensive investigation, it was demonstrated that the calendering method is a significant improvement over existing methods, as it allows for the successful production of high-quality hybrid prepregs. In addition, this method is simpler and thus less expensive than a direct mingling process, so if successful, this can form an efficient alternative to the desired fiber by fiber hybridization that is theoretically shown to provide best mechanical results.

Chapter 7 Microstructural evaluation and mechanical performance of hybrid composites

This chapter aims to highlight the significance of new microstructural analysis techniques specifically developed for examining hybrid thin-ply composite materials. It discusses how microstructural analysis was utilized to ensure the quality equivalence of the newly developed hybrid prepregs with conventional non-hybrid (single fiber) prepregs and to evaluate the efficiency of different industrial hybrid prepreg manufacturing processes, by assessing their impact on the microstructural features of the developed thin-ply hybrid composites. Additionally, an overview of the key mechanical properties, of interyarn: tow-by-tow and intrayarn: fiber-by-fiber calendered hybrid thin-ply composites, that are part of a broad collaborative hybrid materials testing campaign, is presented.

Finally, this chapter explores the possible correlations between the hybrid manufacturing routes, the final resulting hybrid microstructural profile, and the effects on the mechanical performance of the hybrid thin-ply composites to propose optimization strategies for hybrid thin-ply manufacturing methods.

7.1 Evaluation of the microstructural profile of composite UD laminates manufactured with hybrid thin-ply prepreg materials

The primary objective of the current study was to ensure that all the fiber hybridization processes, involving the newly developed or state of the art processing methods, as well as the newly developed equipment and production stages, did not compromise the quality of the produced hybrid thin-ply prepregs and later the hybrid composite laminates and parts.

This research work focuses primarily on the hybridization of two different grades of carbon fiber, the low-strain LS: Mitsubishi HR40 and the high-strain HS: Mitsubishi 34-700. However, the first hybridization attempts for the manufacturing of interyarn tow-by-tow and intrayarn fiber-by-fiber prepregs and composite laminates were performed using E-glass and carbon fibers, as shown in Figure 7.1. During the initial stages of the project, the primary objective was first to achieve the targeted hybrid architectures of interyarn tow-by-tow and intrayarn fiber-by-fiber and the color difference between glass and carbon helped with the initial assessments

To reveal the microstructural profile of the new types of hybrids and verify their quality equivalence with non-hybrid prepregs, unidirectional multilayer laminates were manufactured for microstructural analysis. These laminates encompassed all the different types and ratios, as well as the different hybridization patterns corresponding to each newly developed hybrid interyarn and intrayarn-calendered prepreg in this research.

The hybrid prepregs were stacked to form test plates measuring 120 mm x 120 mm, with an approximate thickness of 2 mm. The thickness was carefully chosen to capture any potential variations, as depicted in Figure 7.4 and adhered to the guidelines or widely recognized mechanical testing standards regarding the sample thickness. The curing of the test laminates and the preparation of micrography samples followed the optimized composite manufacturing and sample preparation processes outlined in detail in Chapter 3. To enable observation over a larger surface area, the stitching feature of the Keyence digital optical microscope VHX-6000 was utilized. This feature allowed consecutive observations to be combined and stitched within the Keyence native image processing software, resulting in high-resolution images of the thin-ply hybrid composite cross-sections. The microscope’s depth-of-field algorithm was employed to determine the height values of specific pixels in the image. By translating the microscope stage in the height direction while capturing images, the algorithm identified the focus point of each pixel. This information, combined with the stage positions, enabled the deduction of the height values of the in-focus pixels, providing valuable insights into the three-dimensional microstructural characteristic of the hybrid materials, and was employed for void content measurements and the quality screening of the laminates.

7.1.1 Microstructural profile of tow-level hybrids

The optical observations conducted on interyarn tow-by-tow thin-ply composite materials confirmed the high quality of this novel hybrid material type. The successful manufacturing of the hybrid prepregs was validated based on various quality criteria, including the uniformity of fiber dispersion, limited resin bleed, and consistent thickness across different tows.

Of particular interest were the regions where different tows intersected within a single layer of prepregs. These areas had never been studied before and could potentially reveal manufacturing issues or fiber incompatibilities. However, upon careful examination of both the glass-carbon Figure 7.1 and carbon-carbon Figure 7.2 tow-by-tow hybrid micrographs, no visible manufacturing-related issues were observed. By employing the depth of field feature of the microscope that was manually targeted on the few visible darker spots of the cross-sections, it was ensured that they were only surfaces, so it can be confirmed that no visible porosity was observed at this scale for both glass-carbon and carbon-carbon tow-level hybrids.

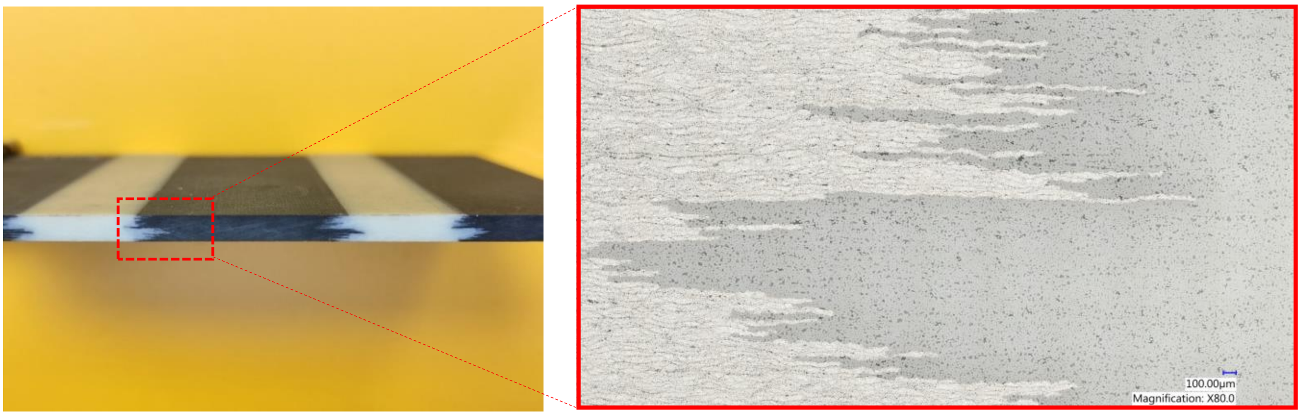


Figure 7.1 Micrograph of HS40 40% - Eglass (60%) tow-by-tow hybrid 54 gsm. The microstructural profile of the laminate reveals high quality at the boarder of the two different tows.

a b

Figure 7.2 a) Tow-by-tow carbon HR40-carbon 34-700 hybrid unidirectional laminate, the borders of the different tows are marked with white lines b) Micrograph of a carbon-carbon tow-by-tow hybrid that reveals good quality at the borders of the different tows.

Apart from the examination at the micro-scale, the materials were also examined macroscopically during the various handling and processing stages that were required for the manufacturing of composite hybrid thin-ply testing laminates. Glass and carbon were initially selected as model materials so as to visually observe the effects of the processing parameters (tension, production line speed, position of the tows, intensity of spreading method) on the quality of hybrid thin-ply composites, during first trial and error tests on the line. As the objective was to manufacture laminates with clear and defined borders between the dissimilar tows, any rolling of fiber, as seen in Figure 7.3, to the neighboring tow, that was interrupting this clear pattern, was considered an unwanted effect. This "rolling" of fibers has been reported in the bibliography as "barreling effect" and it is attributed to a resin flow-induced in-place displacement of the fibrous network during processing resulting from the high pressures applied on the laminate during the consolidation phases of the manufacturing [130] [131].

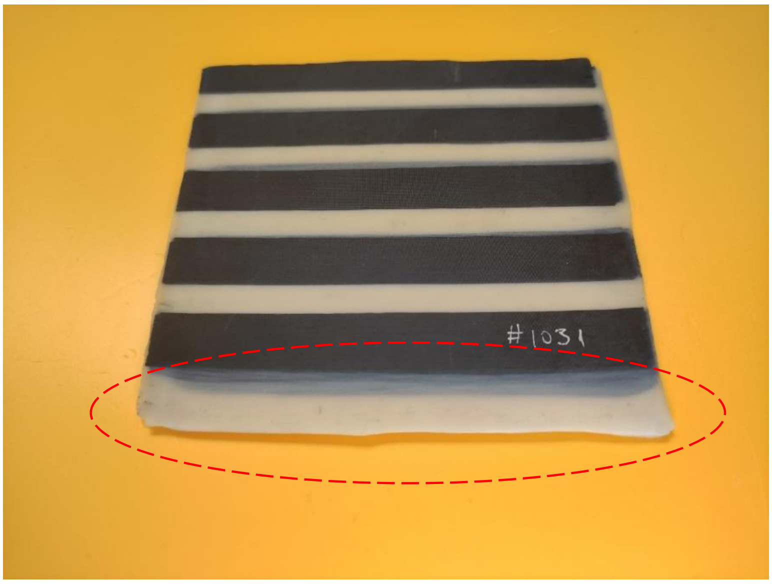


Figure 7.3 Barrelling of carbon fibers to the neighboring glass tow

These observations guided the optimization of vacuum bag assemblies and curing procedures to achieve homogeneous high-quality hybrid laminates as presented in Chapter 3. A reduction of the barrelling effect occurring in unidirectional laminates manufactured with tow-by-tow level hybrid prepregs that were achieved by lowering the curing pressure and using aluminum spacers during curing to reduce the resin flow and restrict the displacement of the fibers can be seen in Figure 7.4.

Figure 7.4 Reduction of the barrelling effect with optimized curing methods that reduced the in-plane movement of the fibrous network to neighboring tows.

Ensuring the quality equivalence of all newly developed interyarn and intrayarn hybrid thin-ply prepregs, regardless of the fiber combination and mixing ratio, to that of non-hybrid single-fiber thin-ply prepregs originally produced by NTPT, was a critical objective of this research. The optimization of manufacturing processes for thin-ply materials can be time-consuming and costly. Any deviation from achieving the desired high-quality equivalence to non-hybrid prepregs would have led to delays and necessitated a reassessment of the initial project objectives. Therefore, close attention was paid to ensuring that the new type of materials could be processed using similar methods as the conventional prepregs, thereby avoiding the need for developing entirely new processes.

7.1.2 Microstructural analysis and development of fiber-level hybrids

The manufacturing process of intrayarn fiber-by-fiber hybrid prepregs and composite laminates underwent several developmental stages as well, to attain the required material quality for subsequent analysis and applications. Initially, a laboratory-scale calendering apparatus was utilized to attempt fiber-level hybridization as described in Chapter 6. While the calendering process showed promising performance in terms of fiber mixing even from the early stages of the research the overall quality of composite laminates manufactured with the preliminary calendered hybrid prepregs did not meet basic homogeneity and low void content quality criteria. As seen in the micrograph Figure 7.5, which was obtained from a cross-section of cross-ply laminate manufactured with calendered glass-carbon hybrid thin-ply prepregs, the void content is extensive, rendering the material unsuitable for further analysis and testing. This high void content of the calendered thin-ply hybrid could only be attributed to a material degradation caused by the calendering process, as intermediate thin-ply glass and carbon thin-ply prepregs that were used for the process had been tested separately and were below the 1.5% void content that was been set as the higher limit by NTPT.

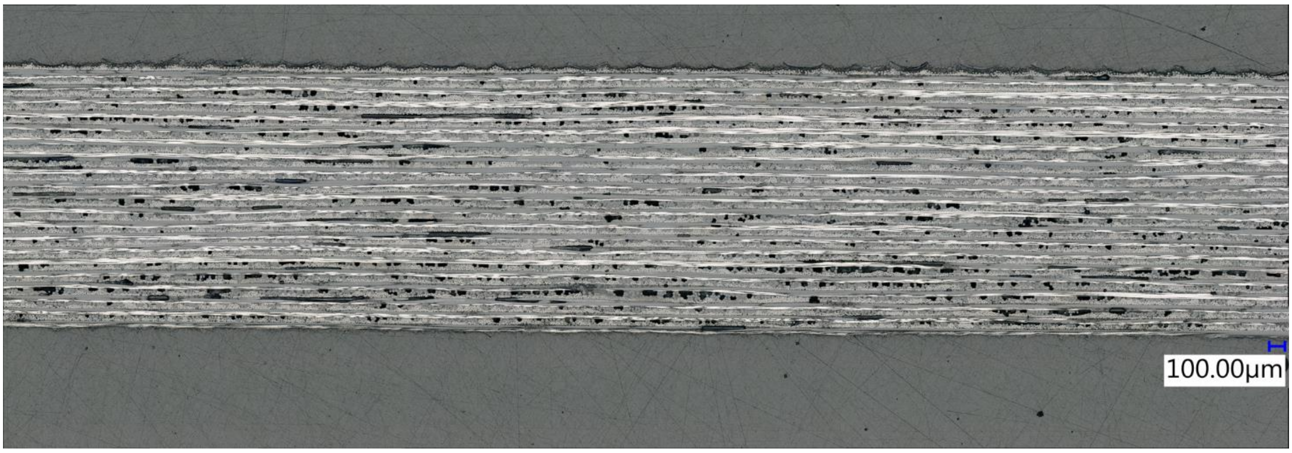


Figure 7.5 Micrograph of a cross-ply laminate manufactured with glass-carbon calendered thin-ply prepregs. The darker areas are porosity due to manufacturing instabilities.

The root cause of this problem was identified to be the uncontrolled high pressure that was applied to the thin prepreg by the calender rolls of the small laboratory-scale calendering apparatus that was employed for the preliminary experiments. This high pressure caused drainage of a significant amount of resin from the intermediate prepregs and although the desired fiber-level hybridization was achieved the resulting calendered hybrid prepreg had an unacceptably low resin content resulting in the formation of large voids in the cured laminate. These preliminary findings were taken into consideration during the subsequent development stages of the calendering process highlighting that low resin content prepregs may be unsuitable as intermediate materials and that the pressure of the rolls had to be adjusted precisely to balance the trade-off between fiber mixing and materials degradation.

After several optimization loops that were carried out by alternating the calendering parameters and then examining the microstructural characteristics of the resulting hybrid thin-ply composite laminates, the preliminary manufacturing issues were addressed, and the method was stabilized. The micrographs obtained from cross-sections of laminates manufactured with the optimized calendering process, revealed for the first time fiber-level hybridization with an impressive fiber spreading down to a single fiber layer for the larger E-glass fibers as reported in Figure 7.6, without visible porosity at this scale.

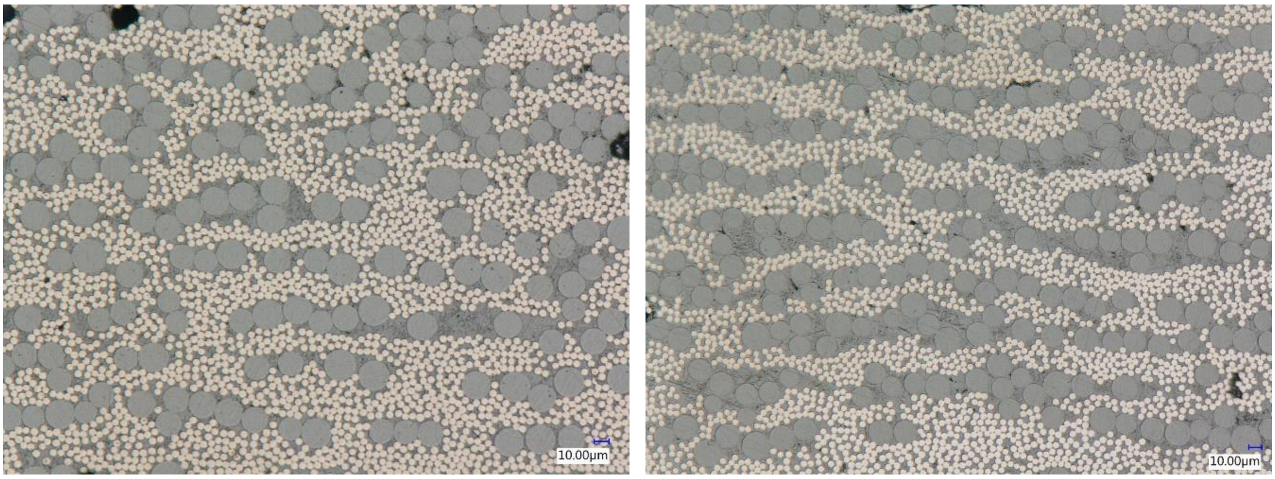
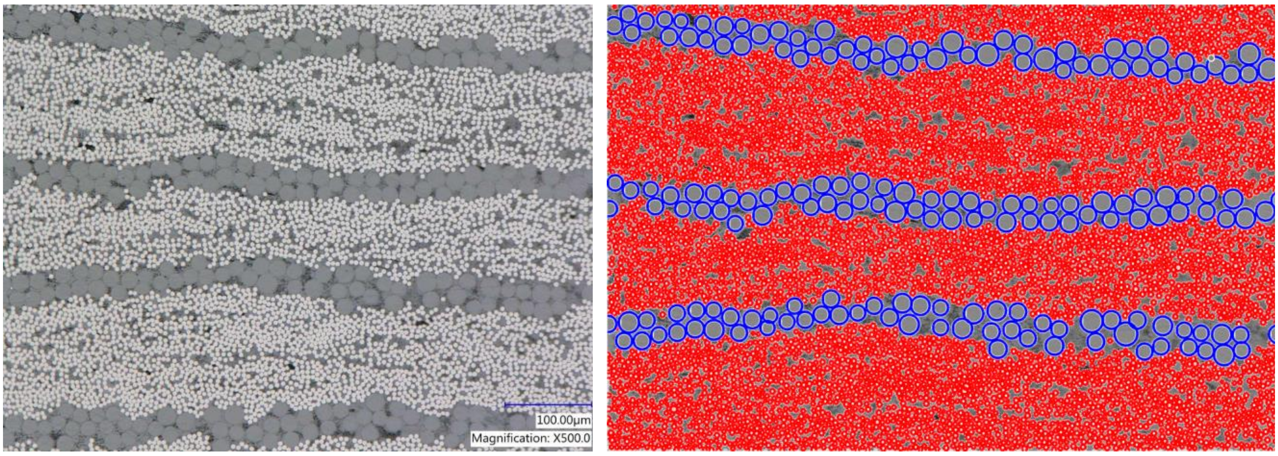


Figure 7.6 Calendered E-glass – carbon intrayarn: fiber-by-fiber fiber obtained with the small-scale experimental calendering set-up.

These microstructural profiles with a fiber-level hybridization together with micrographs obtained from ply-level hybrids produced with ultra-thin prepregs, served as the first imput for the development of a fiber recognition Figure 7.7 and image analysis algorithms Figure 7.8. The objective of these algorithms was to differentiate between the two different carbon and glass fibers and categorize them based on their nearest neighbour. These algorithms served as the foundation for the development of fiber recognition and image analysis tools that were specially tailored for the requirement of the current research project and enabling the accurate distinction between different grades of carbon fiber.



a b

Figure 7.7 a) Micrography of a ply-level glass-carbon hybrid employed for the development of the preliminary fiber recognition tools. b) recognition and separation of each fiber type based on their physical characteristics.

As reported in Figure 7.8 the preliminary microstructural analysis tool could detect fibers and separate them according to the type of its nearest neighbour. As an example, the analysis of the micrograph of Figure 7.7 revealed 6066: carbon-carbon, 121: carbon-glass, 99: glass-glass and 79: glass-carbon neighbouring fibers.

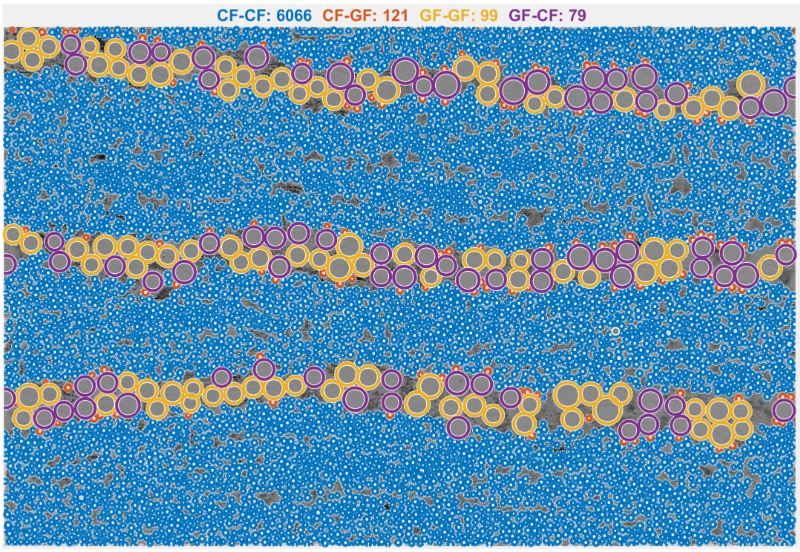


Figure 7.8 Microstructural analysis on a ply-level carbon-glass fiber thin-ply hybrid composite laminate. The fiber are categorized based on their nearest neighbor.

7.2 Microstructural analysis for the evaluation of the calendering process

The microstructural analysis techniques developed for this work were also the main tool for assessing the efficiency of the novel thin-ply hybridization method, the calendering process, as a method for achieving consistent intrayarn fiber-by-fiber hybrid thin-ply prepregs. As discussed in Chapter 6 the fiber-level intrayarn thin-ply prepregs were manufactured by calendering two intermediate very thin-ply non-hybrid low strain LS: HR40 (15 gsm) and high strain HS: 34-700 (30 gsm or 60 gsm), with a width of 300mm in the industrial calendering apparatus that was specifically set-up for this research project in the facilities of NTPT in Poland. For each combination of thin prepregs (HR40 15 gsm + 34-700 30 gsm), (HR40 15 gsm + 34-700 60 gsm) two different pressure settings were applied namely “HP: high pressure” and “LP: low pressure”. Plates were produced following the procedure described previously for the evaluation on the interyarn: tow-by-tow hybrid in section 7.1.1. Additionally, baseline (non-calendered) test laminates were produced by stacking equivalent laminate ply-by-ply. The key objectives were to identify if this method was capable of producing fiber-level intrayarn hybrids using two different grades of fibers, then to quantify the level of fiber comingling that could be achieved while in parallel identifying any quality inconsistencies or damage introduced by the calendering process.

<u>Nomenclature</u>

In what follows, the fiber mixing ratios and the geometric characteristics of the thin-ply prepregs to produce were evaluated by Guillaume Broggi, also working for HyFiSyn project, for translaminar toughness improvement evaluation. Fiber ratio were established to cover several areas of damage mode maps proposed by Jalalvand et [77].

The nomenclature is as follows:

- “B” : signifies Baseline non hybrid samples
- “T”: refers to “Tow-by-Tow” (interyarn) hybrids
- “C”: denotes “Calendered”, fiber-by-fiber (intrayarn) hybrids

The following superscript is the low strain Fiber Areal Weight (typically 15 gsm), and the subscript is the high strain Fiber Areal Weight (30 or 60 gsm). The preceding superscript corresponds to the low-strain fiber proportion in the sample.

For Calendered “C” specimens only two an additional code indicates the calendered pressure setting that was employed to for the manufacturing of the thin-ply prepreg: “LP” denotes prepregs manufactured with “Low Pressure”, “HP” refers to samples manufactured with “High Pressure” setting of the calender rolls. Finally, NC: Non-calendered is refers to the baseline of the calendering process.

As an example, $\mathrm{LP}^{33}C_{60}^{15}$ refers to a calendered hybrid prepreg manufactured by calendering a HR40 15 gsm + 34-700 60 gsm at Low pressure and with a low strain (HR40) volume fraction of 33%.

7.3 Microstructural features of calendered thin-ply hybrid composites

In this section, characteristic microstructures obtained from multilayer laminates manufactured with different types of calendered hybrid thin-ply prepregs will be presented. The micrographs that are reported were obtained from multilayer laminates with three different layer stacking sequence: unidirectional (UD) section:7.3.1 , cross-ply (CP) section 7.3.2 , and quasi-isotropic (QI). The decision to examine laminates with these specific stacking sequence aligns with the quality testing protocols of NTPT, which require the examination of all the new types of prepreg to be examined at these three basic stacking configurations that are commonly used in the design of composites by the industry.

7.3.1 Unidirectional fiber-level hybrid thin-ply laminates

The microstructural profile of unidirectional laminates manufactured with calendered hybrid thin-ply prepregs manufactured with was examined to obtain a global overview of this new type of material and this increased level of fiber comingling. The micrographs from unidirectional laminates also served as a raw image input for the microstructural analysis tools that were specifically designed to study the fiber hybridization of different grades of carbon (carbon-carbon thin-ply hybrids). Upon close examination of the unidirectional laminates, a pattern is observed, characterized by a double layer of the same fiber type on specific areas of the cross-section as highlighted by red lines in the micrograph reported in Figure 7.9. It is important to emphasize that this repeated pattern is attributed to the lamination techniques and is not related in any way to the inefficiency of the calendering process. The development and optimization loops of the calendering process required a considerable number of specimens to be produced, therefore the targeted thickness of each specimen was achieved by manually stacking building blocks cut from a larger preform and not ply-by-ply for time consideration. Furthermore, it should be noted that the microstructural analysist tools that were developed for the characterization of the carbon-carbon hybrids can be targeted in areas between those duplicated fiber layers as highlighted by the yellow squares to a allow a more accurate quantitative characterization of the microstructural features of the hybrid composite.

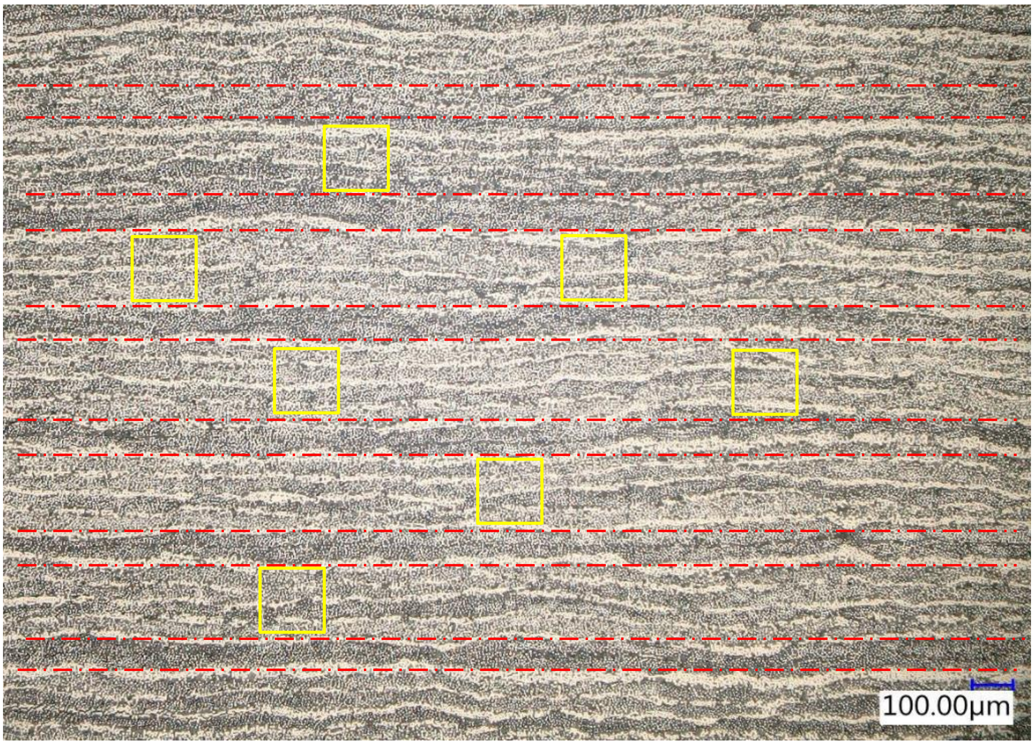


Figure 7.9 Microstructural profile of a unidirectional laminate manufactured with calendered thin-ply prepreg. The red lines highlight double layers of the same fiber that are a result of the preform manufacturing process. The yellow square highlights the areas that are taken into consideration for the microstructural analysis.

The following micrographs (7.10 to 7.13) report characteristic microstructures revealing for the first time the features at this level of fiber hybridization, for two types of stacking: 15 gsm HR40 and 30 gsm 34-700, or 15 gsm HR40 and 60 gsm 34-700. Both types of carbon fibers are distinguished by eye, and the degree of mingling achieved is in the order of 2-3 fiber diameters. No clear difference is observed between the LP and HP calendering process.

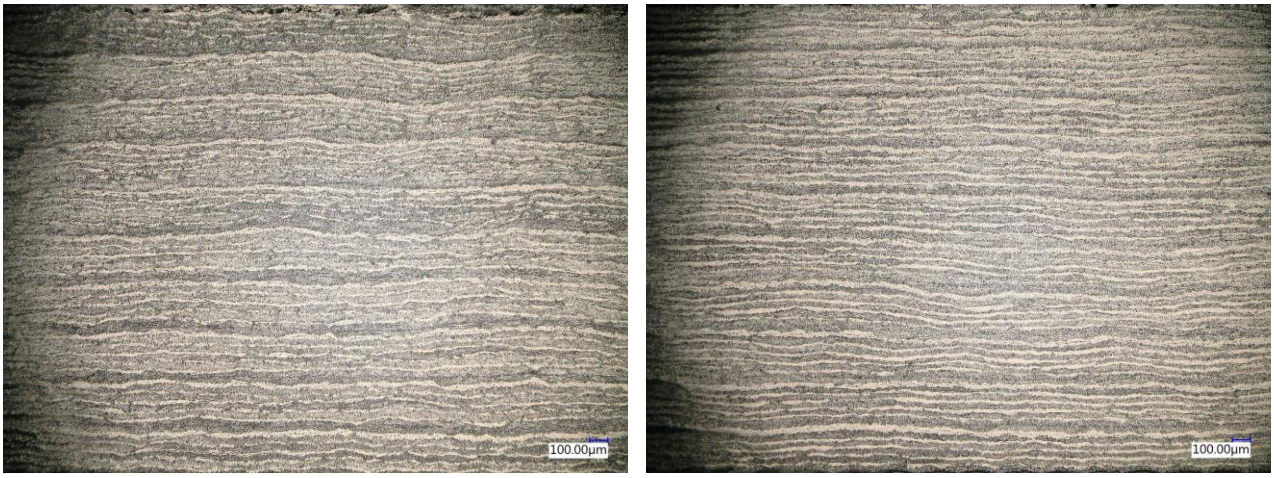


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Figure 7.10 Micrographs of UD laminates manufactured with calendered HR40 15 gsm and 34-700 30gsm at 100X magnification a) $LP^{33}C^{15}_{30}$ b) $HP^{33}C^{15}_{30}$.

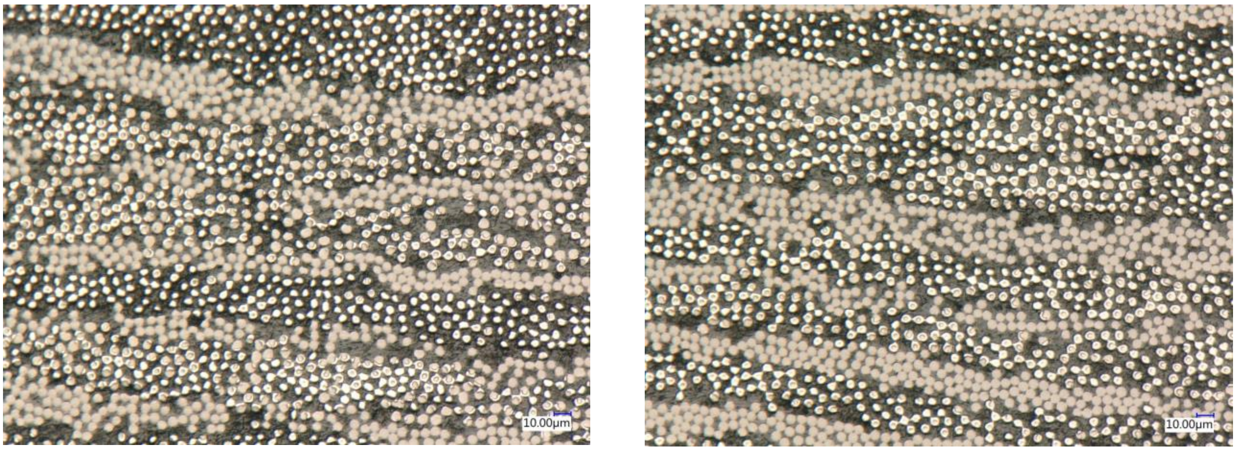

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Figure 7.11 Micrographs of UD laminates manufactured with calendered HR40 15 gsm and 34-700 30 gsm at 1000X magnification a) $LP^{33}C^{15}_{30}$ b) $HP^{33}C^{15}_{30}$.

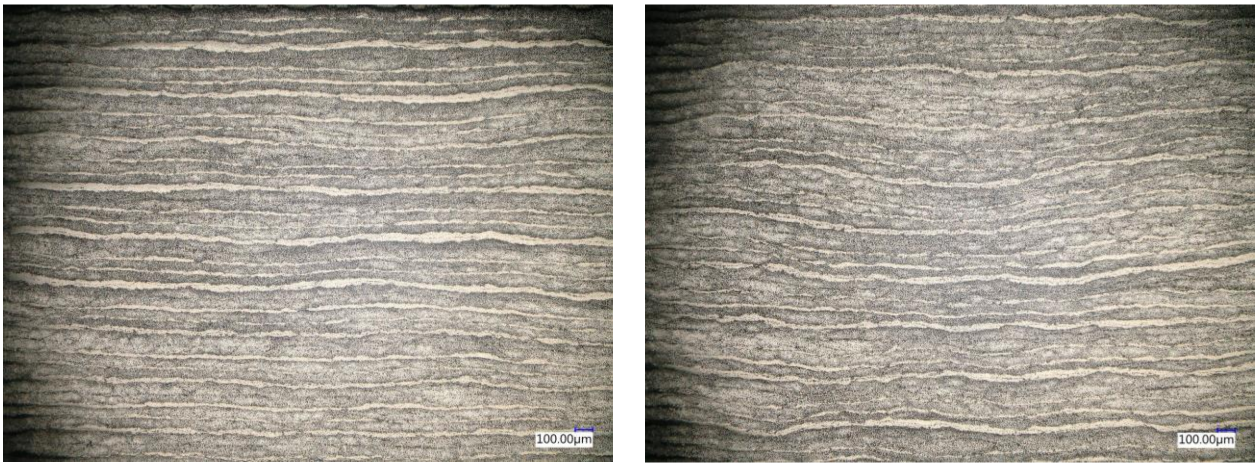

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Figure 7.12 Micrographs of UD laminates manufactured with calendered HR40 15gsm and 34-700 60 gsm at 100X magnification. a) $LP^{20}C_{60}^{15}$ b) $HP^{20}C_{60}^{15}$.

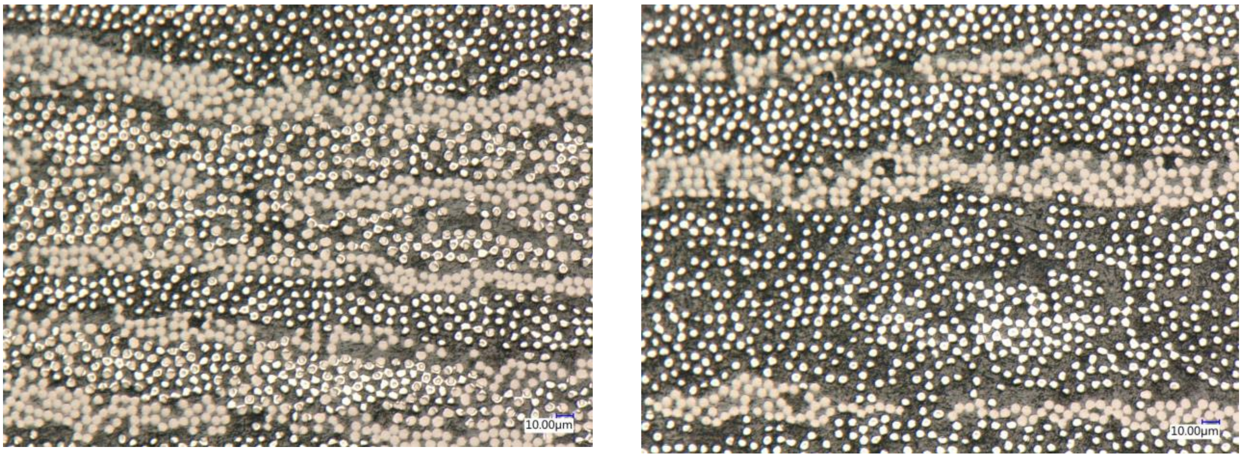

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Figure 7.13 Micrographs of UD laminates manufactured with calendered HR40 15gsm and 34-700 60 gsm at 1000X magnification. a) $LP^{20}C_{60}^{15}$ b) $HP^{20}C_{60}^{15}$.

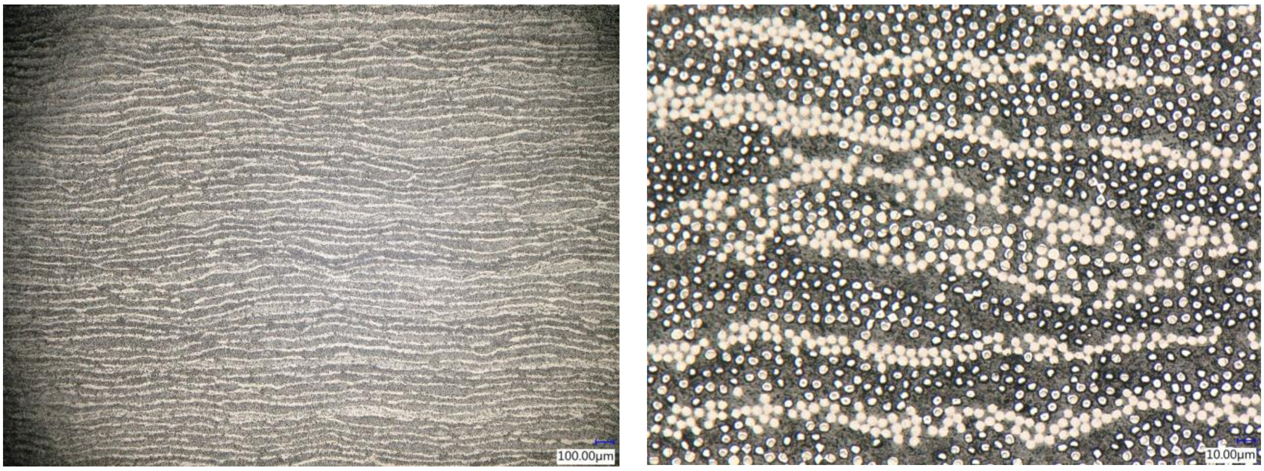

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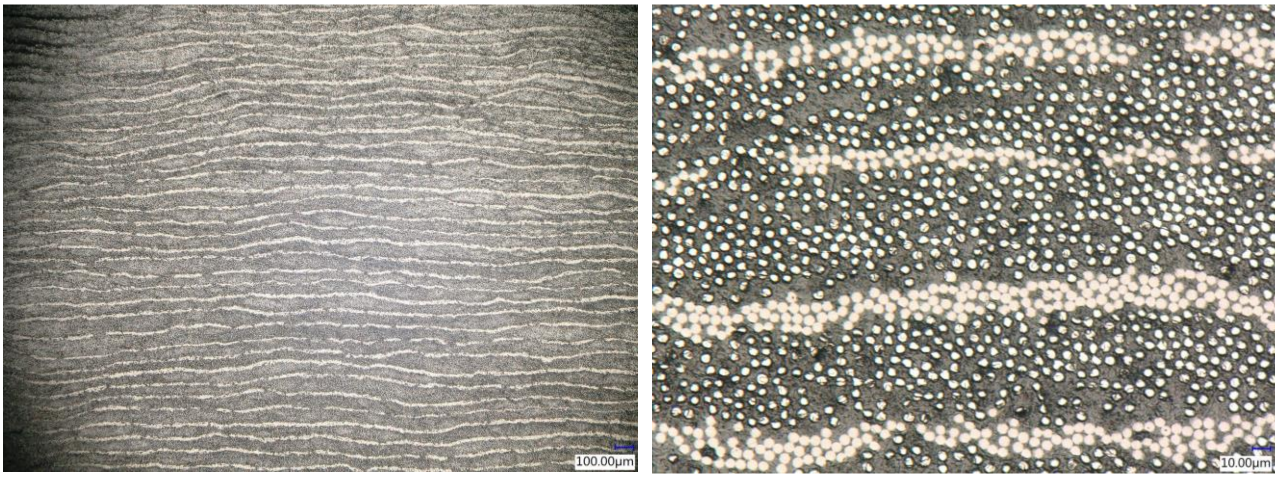
Figure 7.14 Micrographs of UD laminates $N\ {}^{33}C_{30}^{15}$ a)100X b)1000X.



a b

Figure 7.15 $N\ {}^{20}C_{60}^{15}$ a)100X b)1000X.

For comparison, micrographs of the UD laminates manufactured with non-calendered prepregs were also taken and are presented Figure 7.14 and Figure 7.15.

7.3.2 Cross-ply hybrid laminates

The examination of micrographs obtained from cross-ply (CP) laminates manufactured with calendered prepregs was of particular interest, as it allowed for the isolation of the microstructural profile of a single layer of the calendered hybrid prepregs at 0° sandwiched between two 90° layers of the same material, as shown in Figure 7.16, Figure 7.17, Figure 7.18. This isolation provided an opportunity to study the microstructural features of a single layer of hybrid thin-ply calendered material without the influence of transverse movement of the fibrous network, which can occur in unidirectional laminates during the debulking and curing process of manufacturing, although special attention has been given to minimize such movements with the use of a spacer and an optimized curing set-up and vacuum bag assembly, as presented in Chapter 3. Additionally, cross-ply stacking sequence was of particular interest to Guillaume Broggi, also working under the HyFiSyn project, for the manufacturing of compact tension laminates and the study of hybridization as a mechanism to improve translaminar toughness of thin-ply materials [77]. Therefore, we had to verify that cross-ply laminates manufactured with calendered hybrid thin-ply materials met the quality criteria of homogeneity and low void content before supplying the prepreg rolls to the partners of the project for further analysis and testing.

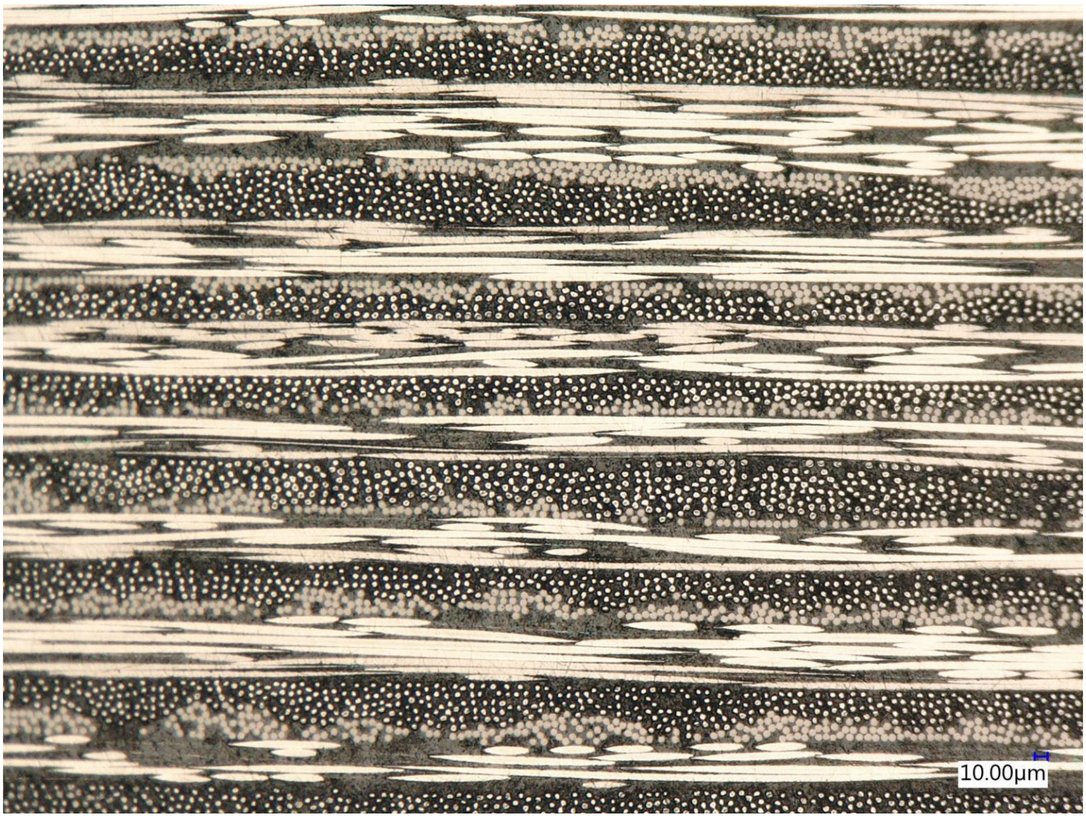


Figure 7.16 Microstructural profile of a cross-ply laminate manufactures with calendered thin-ply prepreg $HP^{33}C_{30}^{15}$ (15 gsm HR40 + 30 gsm 34-700 high pressure) at 1000X magnification.

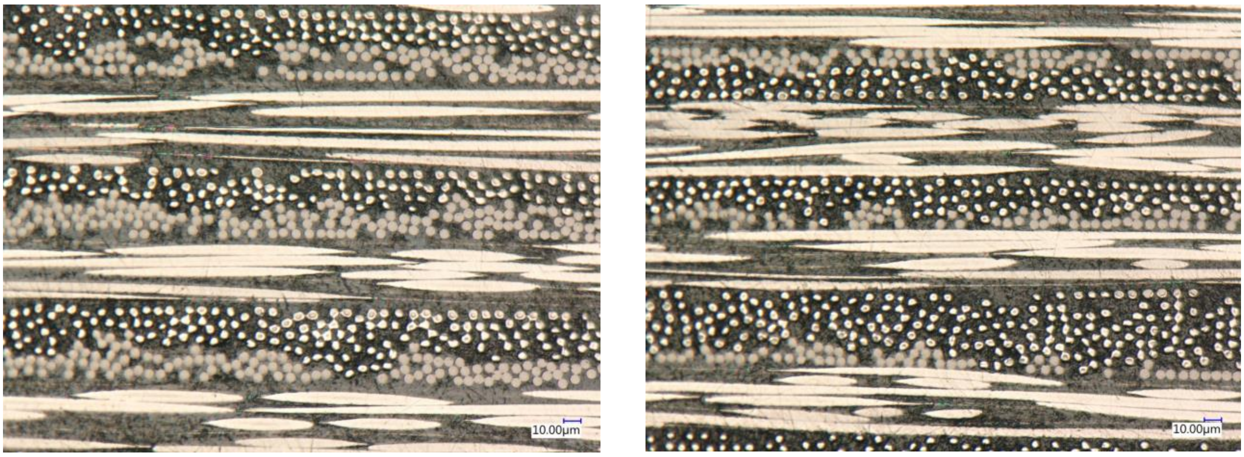


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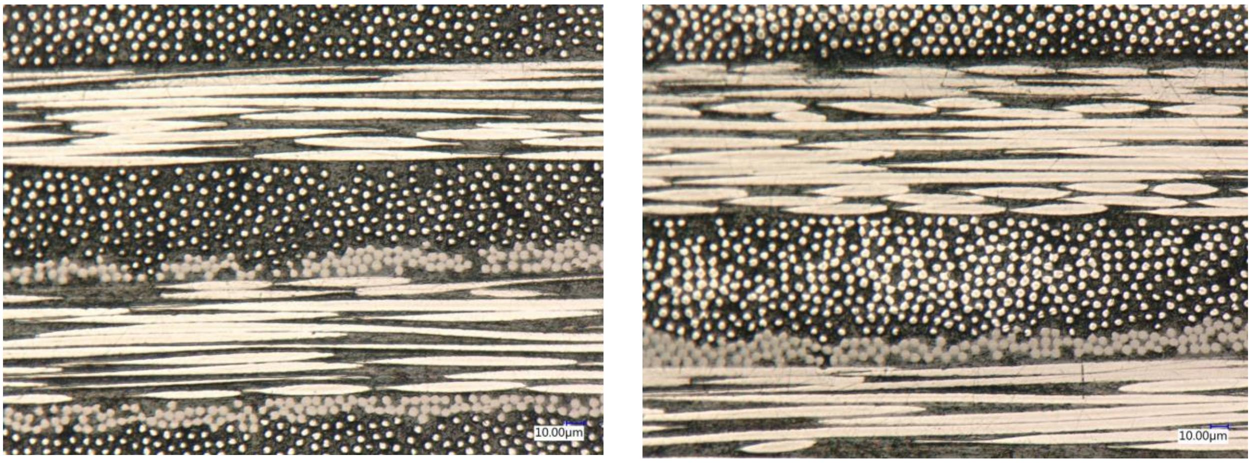
Figure 7.17 Micrographs of cross-ply (CP) laminates manufactured with calendered HR40 15 gsm and 34-700 30 gsm at 1000X magnification a) $LP^{33}C_{30}^{15}$ b) $HP^{33}C_{30}^{15}$.



a b

Figure 7.18 Micrographs of cross-ply (CP) laminates manufactured with calendered HR40 15 gsm and 34-700 60 gsm at 1000X magnification. a) $LP^{20}C_{60}^{15}$ b) $HP^{20}C_{60}^{15}$.

7.4 Evaluation of the influence of the calendering process parameters on the micro-structural profile of hybrid thin-ply composite materials.

This section presents the results from the microstructural analysis method that was developed to quantify the effects of the processing parameters on the overall microstructural profile of hybrid composites produced with calendered thin-ply prepregs. The method and the tools were developed through a collaboration with Dr. Bariș Çağlar and Silvia Gomarasca (Ph.D. student) from Delft University of Technology (TU Delft).

As highlighted in the state-of-art, at the beginning of the current research project, none of the existing micro-structural analysis methods could recognize two different carbon fiber grades in a thin-ply composite cross-section and extract quantitative data based on statistical descriptors in order to allow correlations between the manufacturing route and the resulting microstructural profile. The key features and main working principles of the fiber recognition and microstructural tools as well as and sample preparation procedures have been described in Chapter 3 with the materials and methods.

The volume fractions of the samples are provided in Figure 7.19, separating both types of carbon fibers, as measured by the image analysis presented in Chapter 3. Additional effects arising from debulking cycles are not investigated, as all testing laminates were manufactured following a common debulking process. The resulting general trend shows a slight increase in global fiber volume fraction when the prepreg are calendered. Also, it is worth noting that the proportion of HS versus LS fibers should be similar in the first and second group of three samples, 33% and 20%, which is globally respected, with some variations, which must be attributed to the measurement method.

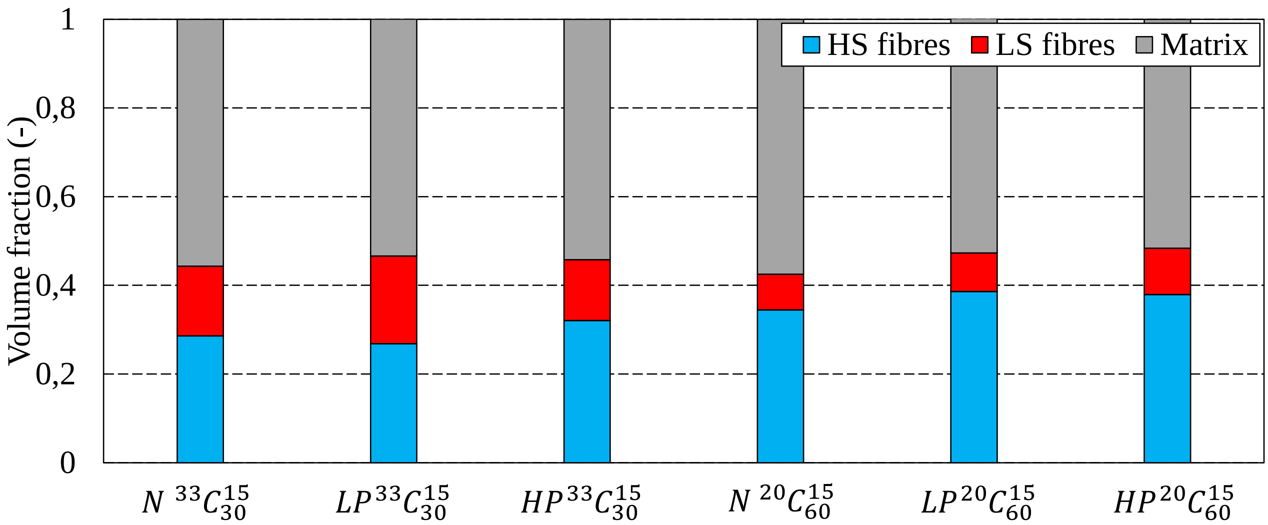


Figure 7.19 Laminate compositions expressed as volume fraction of the individual components.

7.4.1 Areal disorder

The results of the analysis of the areal disorder (AD) are reported in Figure 7.20. The values of areal disorder AD for the global fiber distributions are similar for all samples and lower than the values determined for either LS: low strain and HS: high strain fiber types taken individually. A higher value of global areal disorder AD is observed for the Non-Calendered (NC) samples compared to Calendered (C) samples.

An increase in the value of areal disorder AD for the LS: low strain - fibers (in red) is observed for both increasing calendering pressure and for increasing nominal areal weight of HS: high strain fibers. For the HS fibers (in blue), the value of areal disorder AD has a lower range of variation between samples compared to that for the LS: low strain fibers. A lower value of areal disorder (AD) corresponds to a more regular fiber distribution. For this reason, the areal disorder AD for the global fiber architecture is always lower than for the individual phases, which have a greater variation in tessellation area due to the presence of alternated regions of higher packing density (within each individual layer), and empty regions occupied by the other fiber type. A slightly higher value of global areal disorder AD is observed for Non-Calendered (NC) samples compared to Calendered (C) samples, which might indicate a higher level of overall disorder in the material. Since the increase in HS: high strain fibers nominal areal weight from 30 g/m^2 (gsm) to 60 g/m^2 (gsm) translates into a greater spacing between layers of LS: low strain fibers, the latter phase appears overall less ordered, resulting in an increase in areal disorder AD. For the HS: high strain fibers, the variability between samples can be appreciated less over this range of compositions.

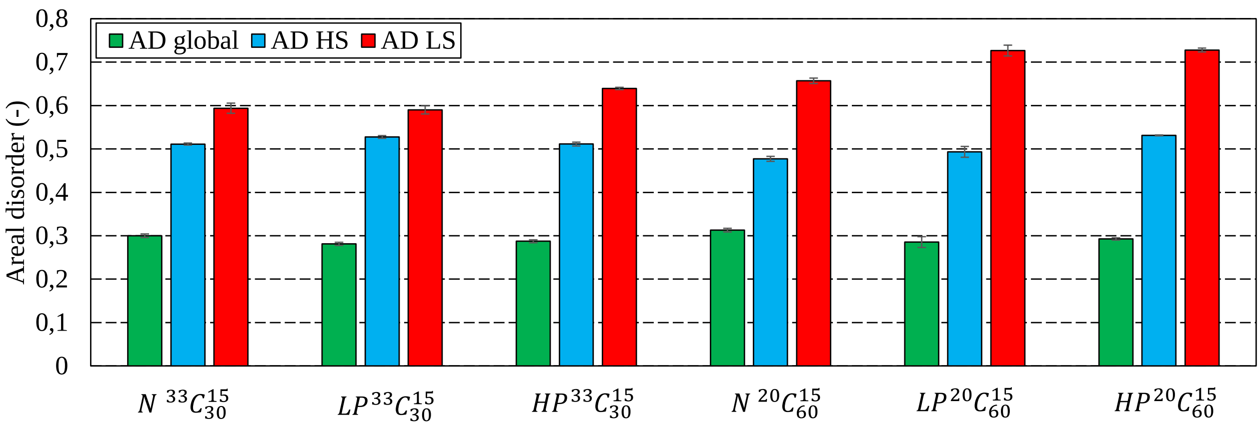


Figure 7.20 Areal disorder for the two fiber types (HS in blue and LS in red) and for the global fiber arrangement (in green).

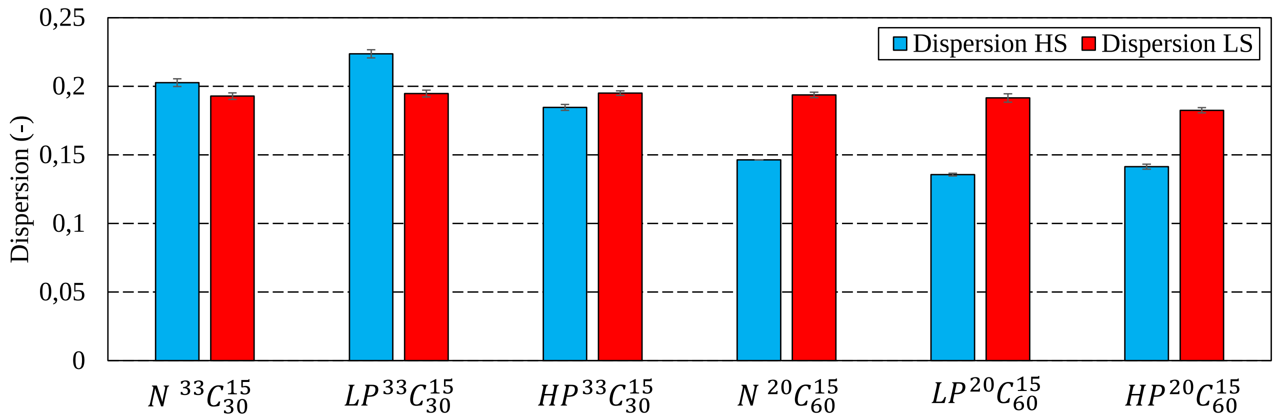


Figure 7.21 Dispersion for the two fiber types (HS in blue and LS in red).

7.4.2 Dispersion

As shown in Figure 7.21, little variation in dispersion is observed between different samples for LS: low strain fibers (in red). Conversely, a general decrease of dispersion for HS: high strain fibres (in blue) is observed with the increase of the nominal HS: high strain fiber areal weight from 30 g/m^2 (gsm) to 60 g/m^2.

Since the areal weight of the LS: low strain fiber layer is similar for all samples, the resulting layer thickness is similar in all samples. For this reason, the distance to the six closest neighbors of the opposite fiber type varies only slightly between samples, leading to small changes in the values of dispersion. Conversely, for an increase in nominal HS: low strain fibers areal weight, the average distance to the six closest LS: low strain fibers will increase. This might have a different effect depending on the effective HS: high strain fiber fraction shown in Figure 7.19 which might determine a variation in the average layer thickness.

7.4.3 Degree of hybridization

The distribution of areal ratios for each sample and for their corresponding random microstructures are shown in Figure 7.22. A higher number of regions containing only HS: high strain fibers is present for increasing HS: high strain fiber nominal areal weight, which is shown by a higher bin count at AR=1. The resulting degree of hybridization shown in Figure 7.23 decreases with increasing HS: high strain fiber nominal areal weight and is highest for the Non-Calendered (NC) samples within each nominal composition. The degree of hybridization was calculated with reference to HS: high strain fibers.

Choosing to refer to LS: low strain fibers instead would have led to a flipped distribution of areal disorder values, and therefore would not have affected the current analysis. A window size equal to six times the nominal fiber diameter was chosen as recommended in the literature [83], however, a different choice of size is expected to impact the result, as the greater the window size, the greater the microstructural homogenization in the resulting AR distribution.

The choice of random microstructures tailored to each sample to match their fibre type composition was considered necessary to generate comparable datasets, due to the variations in fibre type composition shown in Figure 7.19. The greater degree of hybridization for the LS: low strain 15 g/m^2 (gsm) – HS: high strain 30 g/m^2 (gsm) sample series relates to a more homogenous microstructure within the window of observation chosen compared to the LS low strain 15 g/m^2 (gsm) – HS: high strain 60 g/m^2 (gsm) series due to lower spacing between different layer types. Within each sample series, a greater degree of hybridization is observed for non-calendered samples compared to the calendered counterparts.

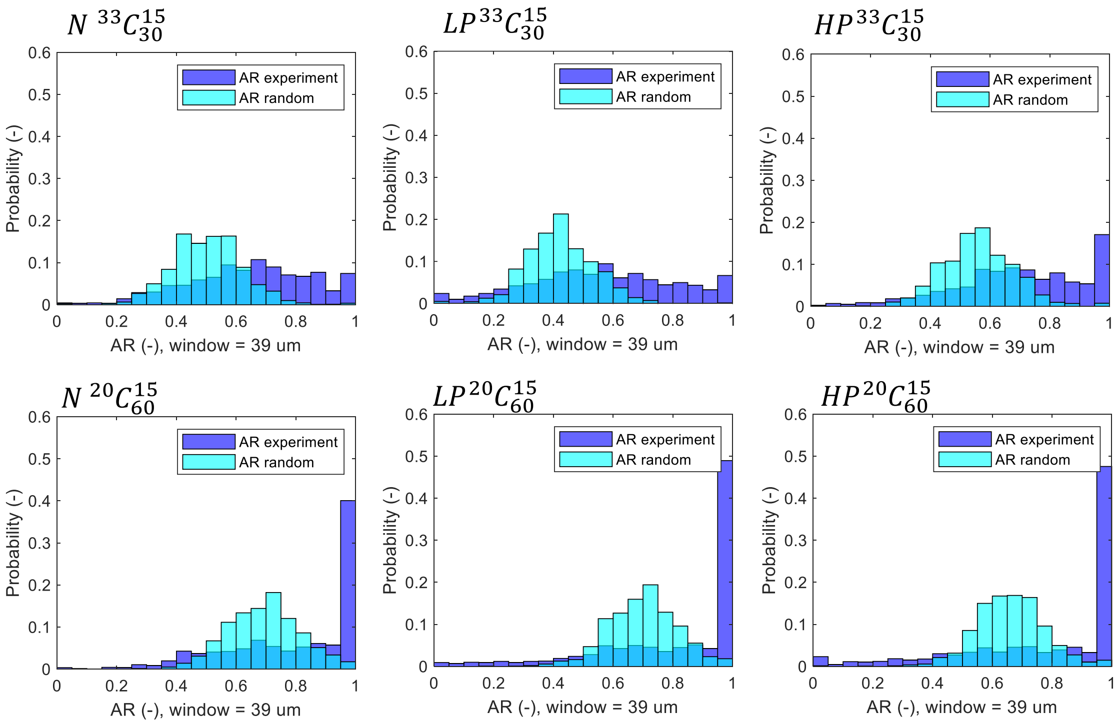


Figure 7.22 Distributions of areal ratio (AR) for the four samples.

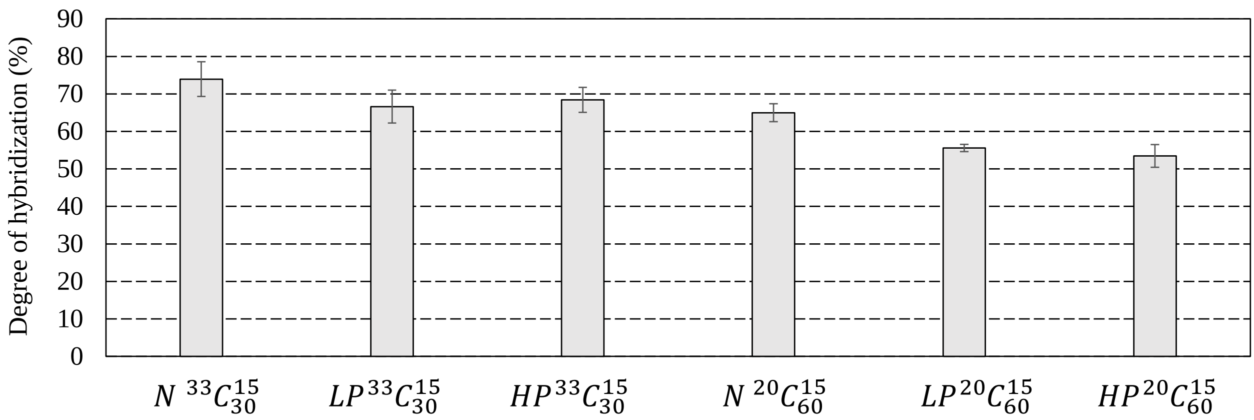


Figure 7.23 Resulting degree of hybridization (H).

7.4.4 Conclusions

Three parameters were used to describe the hybrid microstructure: areal disorder [109], dispersion [110], and degree of hybridization [83]. All parameters were able to capture differences between samples which are linked to size effects related to varying layer thickness. Calendering effects were mostly captured by areal disorder and degree of hybridization. Lower areal disorder in LS: low strain fibers and greater degree of hybridization were encountered for Non-Calendered (NC) samples compared to the High-Pressure Calendered (CHP) counterpart, suggesting greater local fiber intermingling for the former. This result was unexpected a priori, however, a careful observation of the micrographs of the non-calendered composites shows that the pressure applied during production of these very thin plies enables a fairly good level of mingling, and possibly less disorientation of the fibers as the calendered ones. This is true for the UD, however, for cross-ply and Quasi-iso plates, the advantage of pre-mixing by calendering, in terms of prepreg handling and lay-up was considered to still be advantageous. Finally, it is worth noting that the HP and LP settings used in the calendering were set on an industrial machine and restricted due to limited time on real production machines, so this production step would need to be further explored for full assessment of the technique.

7.5 Mechanical characterization overview

One of the significant drawbacks of working with (hybrid) thin-ply prepreg materials is the increased production time that is required for the manufacturing of composite laminates or structures due to the low thickness of the building block (individual ply). Additionally, for ultra-thin ply prepreg formats like 15 g/m^2 (gsm) HR40 used in this study, extra care must be taken during lamination as the material may be more sensitive to handling and may require some additional debulking to facilitate the removal of the backing/carrier paper, resulting in longer specimen production campaigns compared to conventional prepregs.

To maximize the production of testing specimens and the extraction of data for the evaluation of the mechanical performance of the hybrids, certain manufacturing steps and mechanical tests were conducted in collaboration with Guillaume Broggi. Consequently, some of the mechanical testing results are shared. When there is a difference either in the manufacturing or testing approach, it will be highlighted and the impact of these different manufacturing methods on the results will be discussed.

Aluminum tabs were used for the Baseline specimens produced at EPFL, due to the convenience in manufacturing. However, such specimens required waterjet cutting, as the diamond disc saws typically employed for the specimen preparation cannot be used to cut through. The Interyarn: tow-by-tow : “T” and the Intrayarn: fiber-by-fiber: Calendered: C specimens were manufactured with 2 mm thick, $[+45, -45]_{ns}$ carbon composite tabs of 34-700 gsm thin-ply instead of aluminum, which is the typical specimen procedure followed by NTPT, as producing the carbon tabs and preparing specimens out of carbon was consider cheaper and more convenient due to direct access to raw materials and tools. Both aluminum and carbon fibers tabs were found to transmit the load between the grips and the samples efficiently without any noticeable variation in the performance of the specimens or the testing procedure.

7.5.1 Unnotched tensile properties of baseline thin-ply carbon fiber composites

This section presents the experimental results for unnotched tensile UNT mechanical characterization of quasi-isotropic laminates QI laminates manufactured with single fiber (non-hybrid) thin-ply prepreg materials which were used a baseline. The production of the single fiber prepregs was done using the NTPT fiber spreading and impregnation methodologies presented in Chapter 3.

The four QI laminates for the UNT test were manufactured by Guillaume Broggi at EPFL and were tested in collaboration to study the baseline properties of the thin-ply prepreg materials used in this work. One laminate was manufactured with the LS: low-strain: HR40 30 gsm: ${}^{100}B_{0}^{30}$. Three laminates were manufactured with different thickness (60,120,180 gsm) of the high strain 34-700 to evaluate the thin-ply effects: ${}^{0}B_{60}^{0}$, ${}^{0}B_{120}^{0}$, ${}^{0}B_{180}^{0}$.

The stress-strain curves of the materials that were tested as baseline are presented in Figure 7.24. The low-strain prepreg as expected fails around 1% strain, whereas the high-strain baselines exceed 1.5%. The ply effect is also observed, with a strain to failure lower as the ply-thickness increases, as will be discussed later.

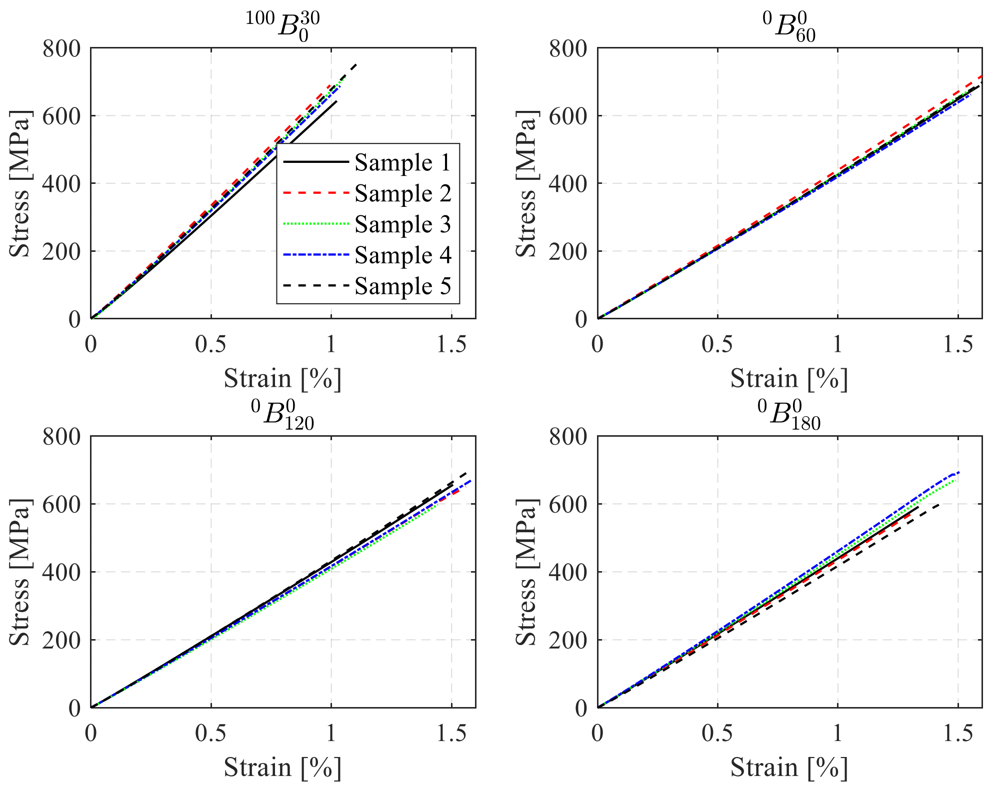


Figure 7.24 Stress-strain curves of the baseline materials.

7.5.2 Unnotched tensile properties of interyarn: tow-by-tow thin-ply composites manufactured with two different grades of carbon

This section presents the experimental results for the for UNT mechanical characterization in QI laminates manufactured of Interyarn: tow-by-tow: "T", fiber-hybrid thin-ply prepregs materials. The production of the Interyarn: tow-by-tow prepreg with controlled characteristics in terms of fiber types and rations, ply thickness – FAW and hybrid pattern was achieved after the development of the tow-level hybrid thin-ply prepreg manufacturing methods presented in Chapter 5.

The three QI laminates that were tested are the: ${}^{31}T^{90}$, ${}^{25}T^{90}$, ${}^{31}T^{45}$.

The stress-strain curves of the materials that were tested as baseline are presented in Figure 7.5.

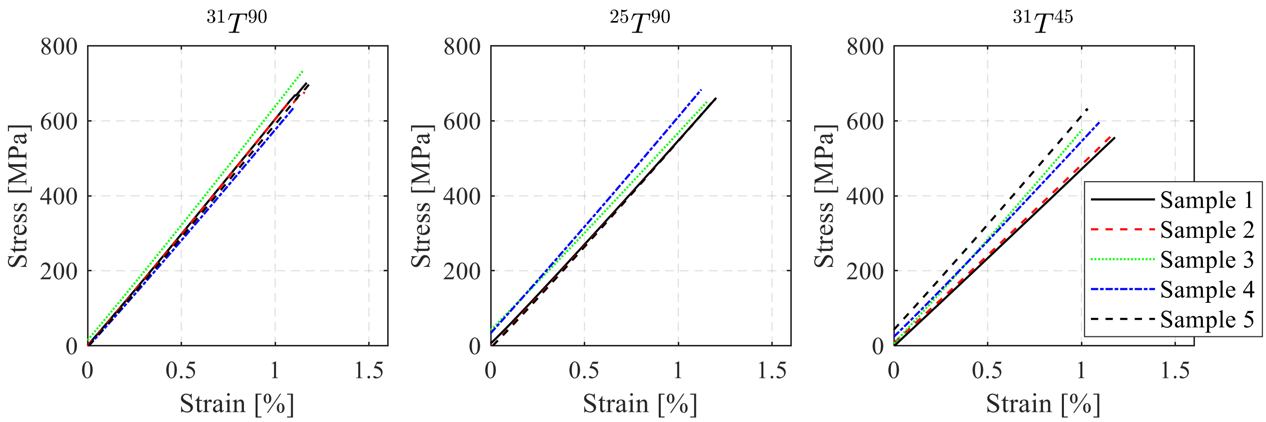


Figure 7.25 Stress-strain curves of Interyarn: tow-by-tow laminates.

7.5.3 Unnotched tensile properties of intrayarn calendered fiber-by-fiber thin-ply composites manufactured with two different grades of carbon

This section presents the experimental results for the for UNT mechanical characterization in QI laminates manufactured of Intrayarn: fiber-by-fiber: Calendered: “C”, fiber-hybrid thin-ply prepreg materials. The production of the Intrayarn: fiber-by-fiber was done by calendering two very thin-plies of HR40 (15gsm) and 34-700 (30 or 60 gsm), with different calendering pressure (high-low pressure) using the calendering processing method (in the direction of the fibers) presented Chapter 6.

The four QI laminates that were tested are the: LP$^{20}C_{60}^{15}$, HP$^{20}C_{60}^{15}$, LP$^{33}C_{30}^{15}$, HP$^{33}C_{30}^{15}$.

The typical stress-strain curves of the calendered hybrid thin-ply prepregs are presented in Figure 7.26.

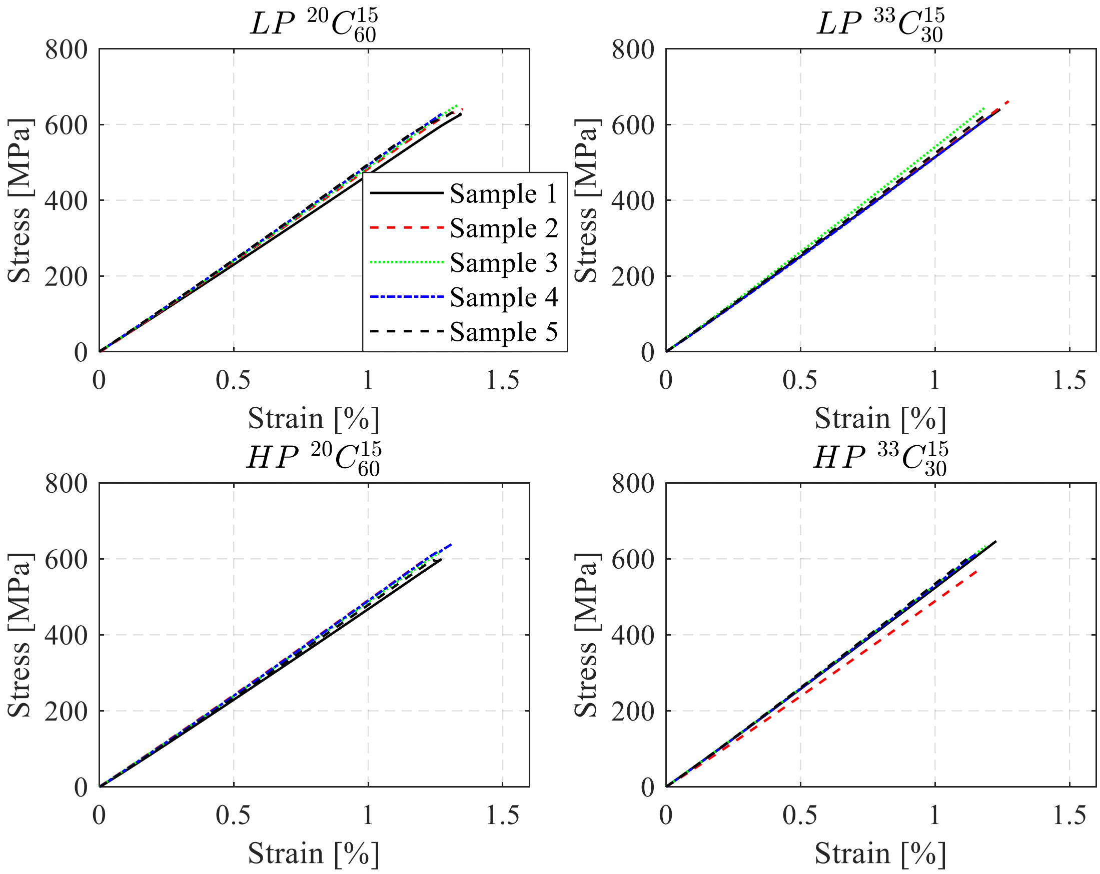


Figure 7.26 Stress-strain curves of calendered materials with different fiber ratios and calendering processing parameters (low-high pressure).

Figure 7.27 illustrates enlarged section of the stress-strain curves of QI specimens manufactured with calendered hybrid thin-ply prepregs. A slight non-linearity before failure can be observed which can be an indication of some level of pseudoductility.

A clear positive thin-ply effect in the failure strain of the baseline 34-700 can be observed in Figure 7.30 with the reduction of the ply thickness. However, the experimental failure stain of 34-700 was measured to be approximately 1.5% and never reached the reported datasheet value of 2.06%. Based on these results G.Broggi updated the initial damage mode maps that were the basis for the design of the calendered materials. Now the ratios are closer to fragmentation zone, this can explain some pseudoductile behaviour and indicated that the properties of the calendered materials can be tuned further to each targeted failure mechanisms.

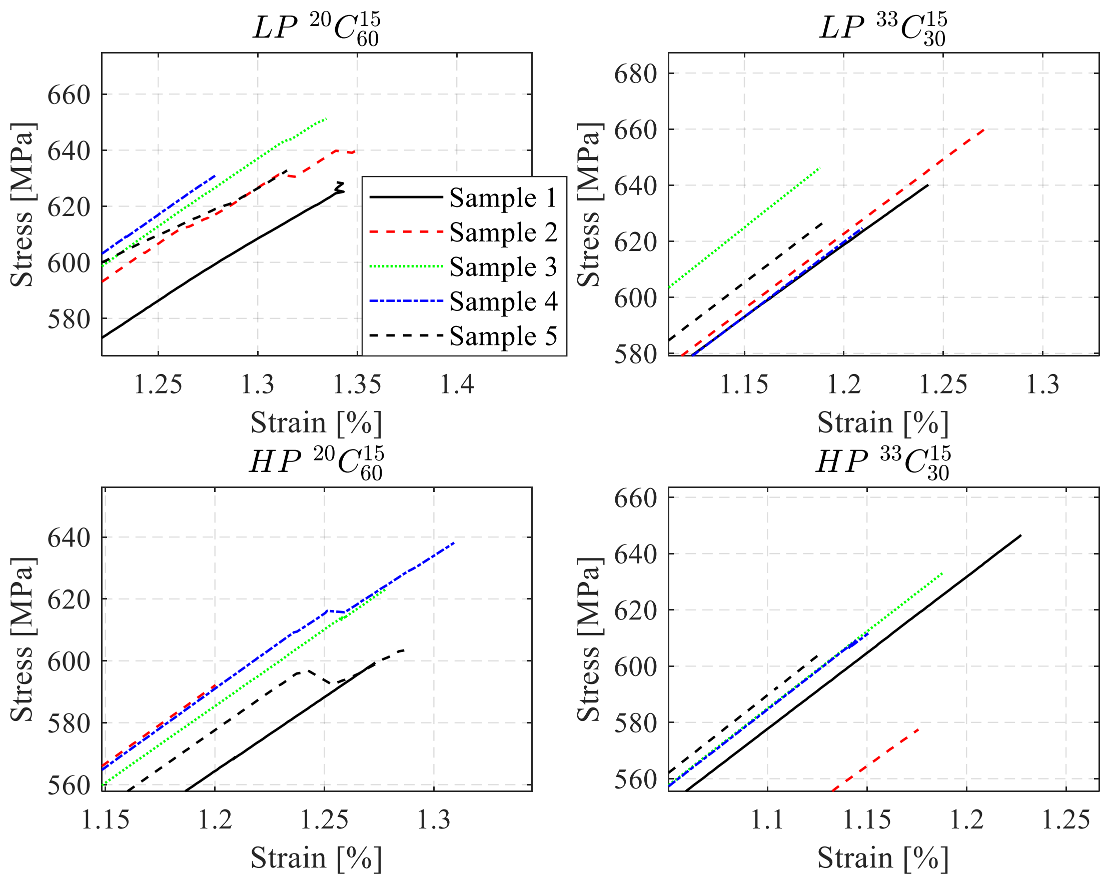


Figure 7.27 Non-linearity before failure for both (high and low pressure) calendered $^{20}C_{60}^{15}$ prepregs.

Figure 7.28 summarizes the Young's modulus of all the composites tested.

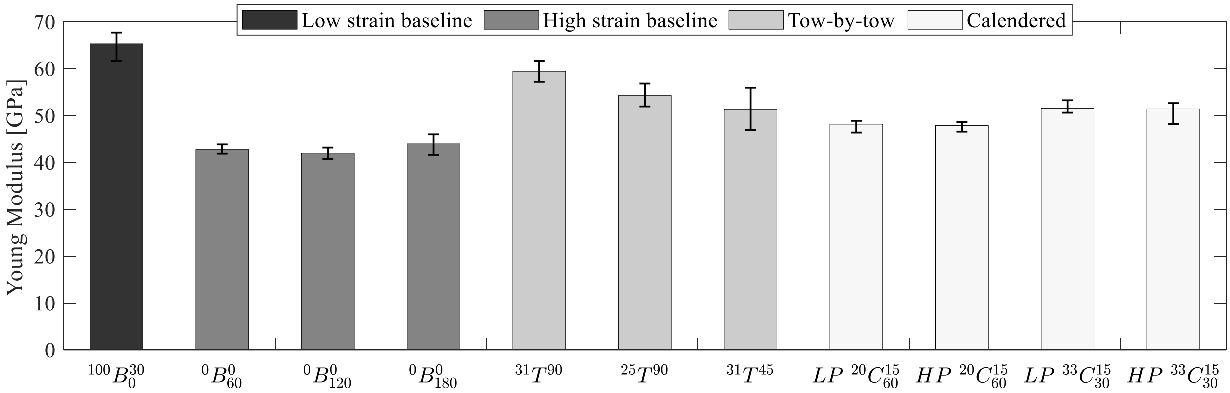


Figure 7.28 Overview on the Young modulus.

As reported in the Figure 7.29, the hybrid laminates strictly follow a rule of mixture as a function of the proportion of low-strain fiber fraction. This indicates that the quality of the prepregs, even after two mixing or calendering is excellent, in terms of fiber quality and fiber orientation.

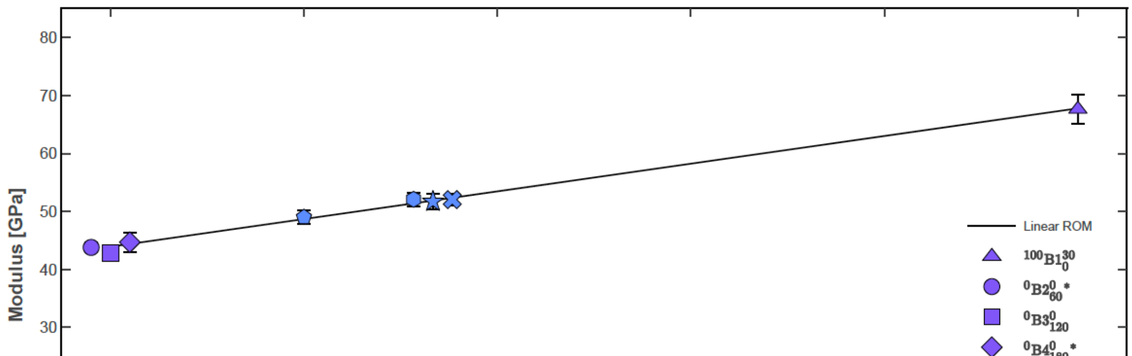


Figure 7.29 Overview of the moduli of the prepregs as a function of the low-strain volume fraction [77].

Figure 7.30 presents an overview of the strain at failure for all composites. Tow by tow hybrids show a rather low failure strain, and large scatter, this is due to the relation between the width of the spread tows as compared to that of the tensile coupon. However, these materials were shown by G. Broggi to be the most promising combinations for translaminar toughness improvement, as parts of the high strain tows bridged the crack and led to increased fracture energy. One of the main limitations of the tow-by-tow process is the impossibility to divide the tows. This limits the tow-by-tow hybridization patterns, as often the low-strain volume fraction indicated by the damage mode maps cannot be achieved in real life. This hybridization method should thus be further optimized. Thinner tows may be used; however, these are expensive. Another possibility would be to propose the addition of a splitting element in the tow-by-tow combs that will increase the controllability and the tuning of the hybridization patterns, since we demonstrated in previous chapters that such a pre-treatment does not greatly affect the fiber strength.

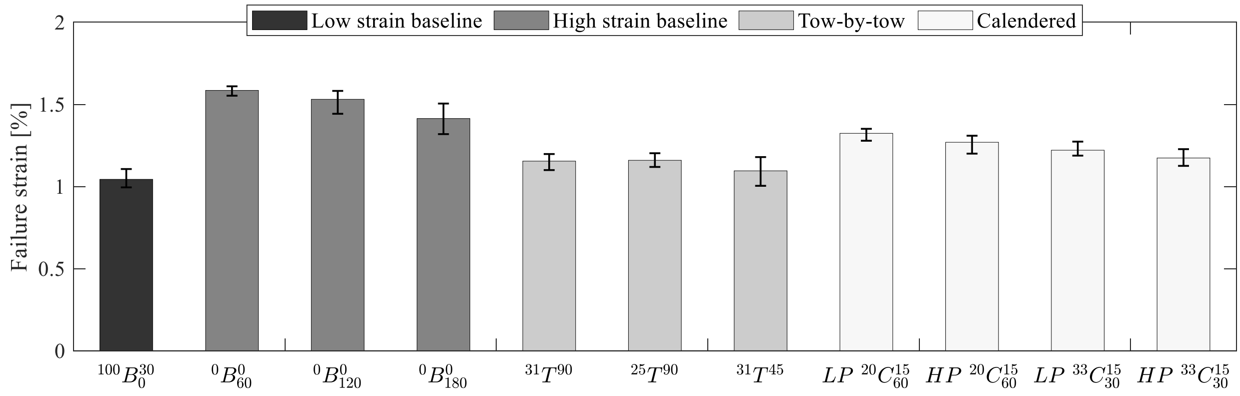


Figure 7.30 Overview of the strain at failure.

In Figure 7.31 an overview the stress at failure for all composites, showing similar trends as for the strain to failure.

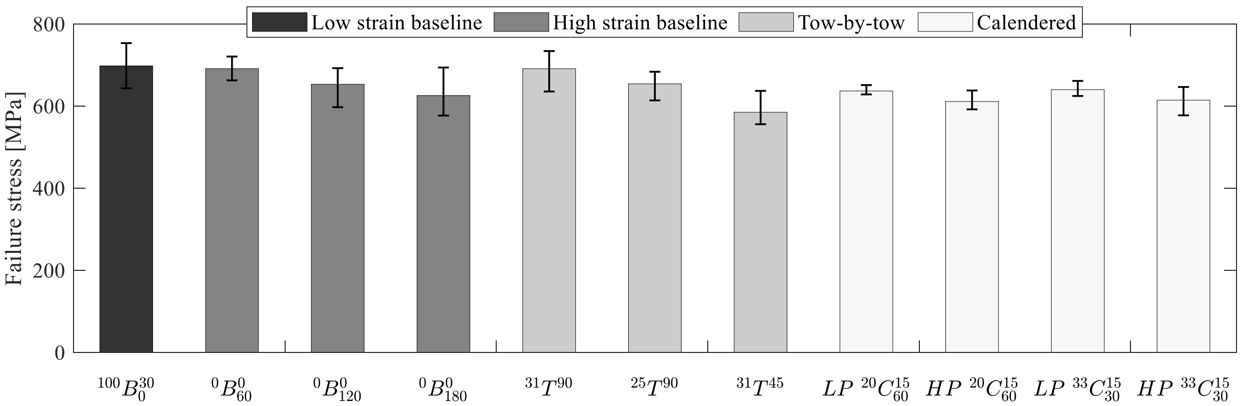


Figure 7.31 Overview of the stress at failure.

The ultimate stress for the tow-by-tow and the calendered composite is similar to that of the low and the high strain baseline. Moreover, the calendered composites show lower dispersion in the ultimate stress compared to both baseline and tow by tow hybrids. The former gives additional credit to the method of calendering and is explained by the fact that controlled fiber mixing that can be achieved in a calendered ply-block. A reduction in stress at failure and strain at failure in the order of 5% is observed with the increase of the calender roller pressure setting from LP: low-pressure to HP: high-pressure. This highlights the need for further investigation of the effects of the calendering parameters on the mechanical performance of the thin-ply prepregs.

7.5.4 Failure mode description

Brittle failure is observed for all the specimens, as shown in Figure 7.32. Since these are all QI specimens, failure of the plies at 45° angle is clearly observed as well. No major difference of failure mode is observed between the sample types, it is also observed that some samples break close to the grips, however most samples broke further away from the grips, often in 2 locations. Observation of the fracture surfaces of the baselines confirm that the low strain baseline fails in a brittle manner, with a sharp and clear fracture surface. The high strain baselines show increasing pull-out and ply delamination as the ply thickness increases, as expected. In the ply-by-ply configurations (from G. Broggi work), as well as calendered ones, an intermediate behaviour is observed, with increased ductility as shown earlier for the calendered samples $^{20}C^{15}_{60}$. The tow-by-tow configurations Figure 7.33 are rather difficult to interpret as the size of the tows was large as compared to the width of the samples, however some ply pull-out and delamination is also observed for these in general.

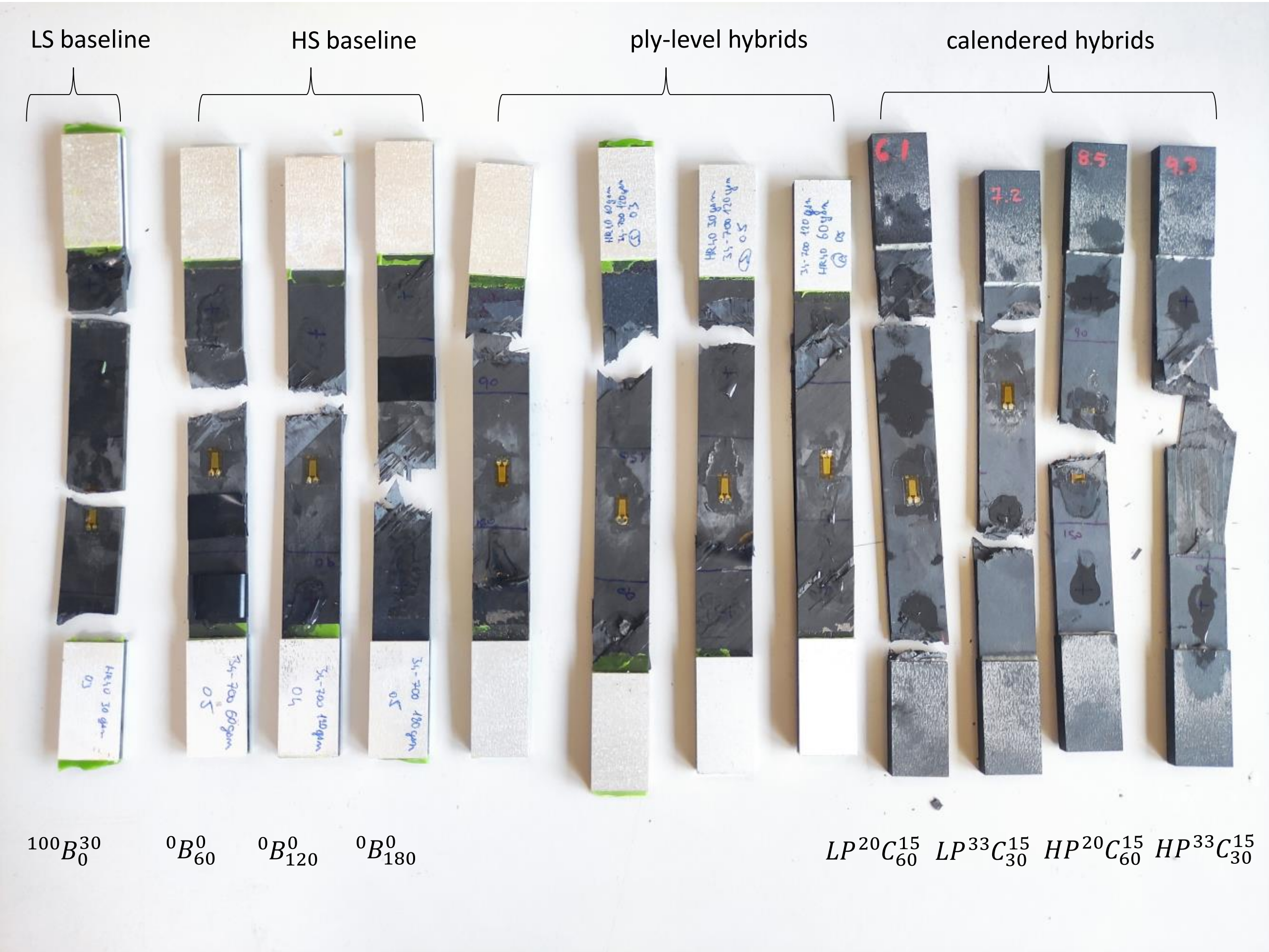


Figure 7.32 Typical examples failed of baseline, ply-level, calendered specimens.

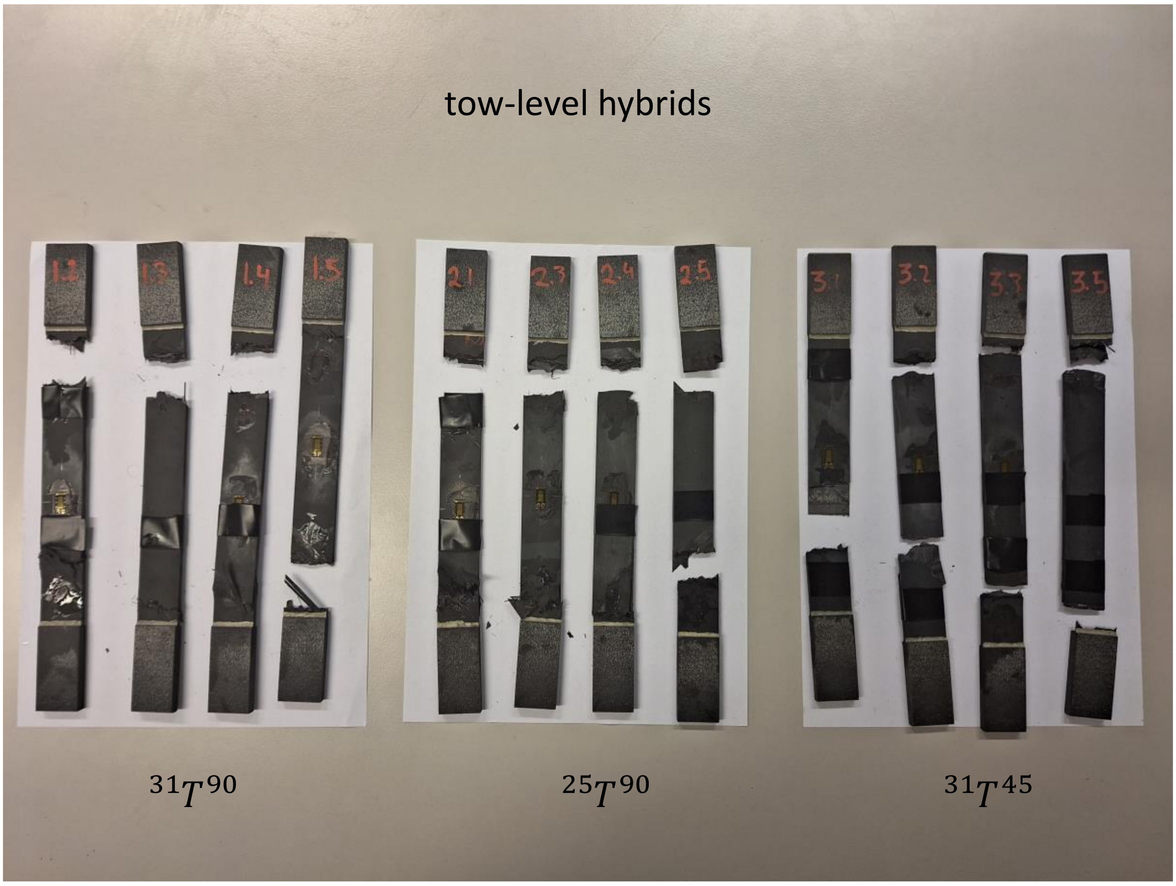


Figure 7.33 Typical examples of failed tow-level specimens.

SEM observations have been carried out on the fracture surfaces at the Interdisciplinary Centre for Electron Microscopy (CIME - EPFL) with a Zeiss GeminiSEM 300. The observations on the calendered specimens reveal good manufacturing quality and some extent of fiber pull-out. The microstructure of the epoxy resin, with the toughening particles that are clustered away from the fibers is also observed. Any effect of high or low calendering pressure cannot be observed at this level of magnification Figure 7.34.

Observations on the fracture surfaces on of tow-level hybrids reveal the delamination that occurred during the fraction Figure 7.35 and also some individual tows of fibers and the different fiber orientations of the QI lamination can be distinguished Figure 7.36.

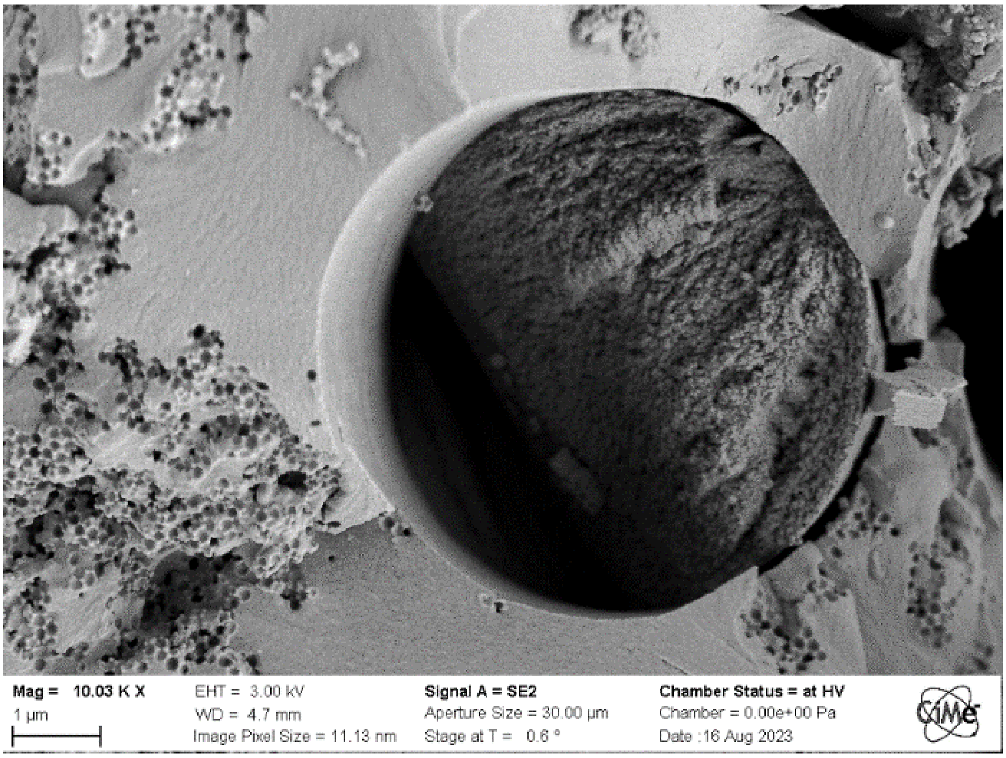


Figure 7.34 SEM image on the fracture surface of calendered LP$^{20}C_{60}^{15}$ specimen.

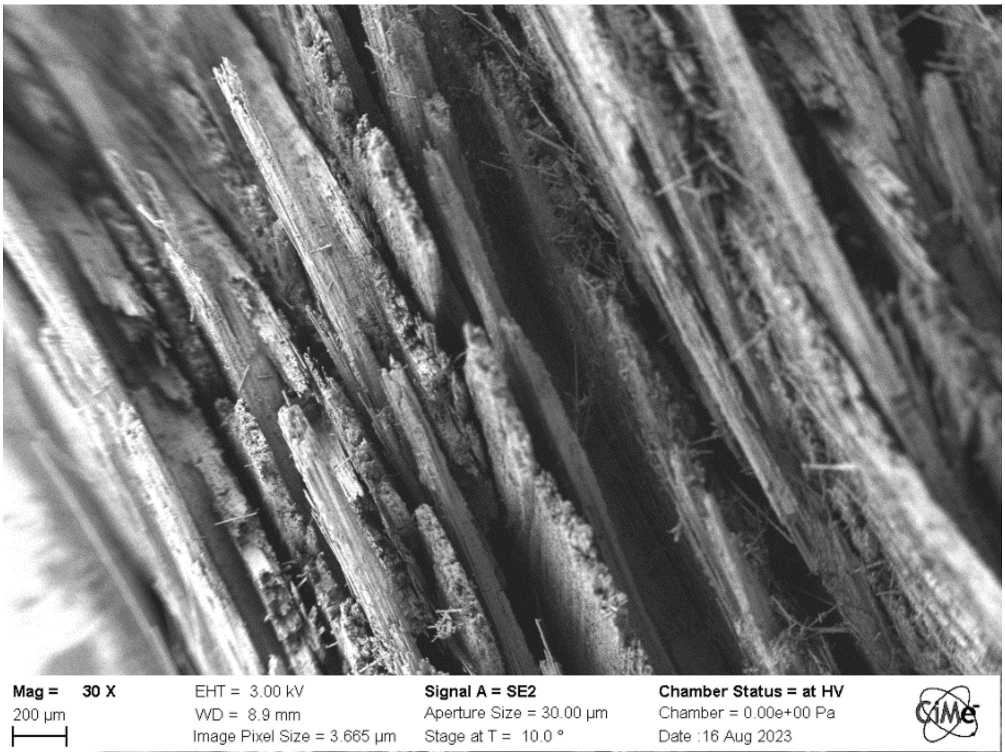


Figure 7.35 SEM image on the fracture surface of tow-level $^{31}T^{45}$ specimen revealing the level of delamination.

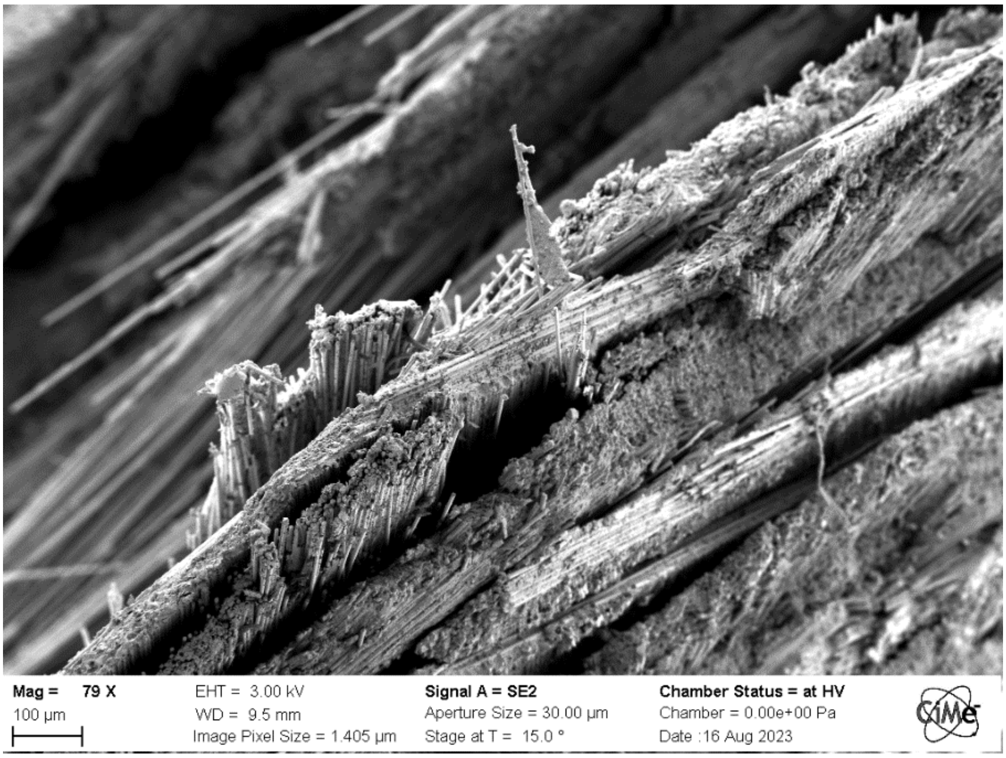


Figure 7.36 SEM image on the fracture surface of a tow-level ${}^{31}T^{45}$ specimen focusing on an individual tow.

Chapter 8 Conclusion and outlook

8.1 Summary and conclusions

This work focused on the development of processing methods for the manufacturing of hybrid thin-ply prepregs at an industrial scale. The existing prepreg manufacturing methods of the composite's manufacturer North Thin Ply Technology (NTPT) were studied and an extensive review of the academic bibliography and technical information from patents sources was conducted to identify critical processing variables and production stages in state-of-the-art prepreg manufacturing that could be better controlled and reveal areas of potential optimization in order to produce hybrid thin-ply prepregs. From a practical standpoint, with the development of new hybrid prepregs, this research enabled the exploration and analysis of the performance improvements in hybrid composite materials with a level of fiber mixing that had never been achieved before.

This work had the unique capability to comprehensively track the entire lifecycle of the materials, starting from the raw materials until the final prepreg product was obtained. This enabled a comprehensive study of the effect of key processing techniques, commonly utilized in the industry, specifically fiber mechanical (pre) spreading and fiber thermal treatment, on the performance of both individual fibers and composite laminates through a research collaboration with DTU on the quality of glass fiber prepregs.

Custom-made equipment was added to the existing R&D prepreg line of North Thin Ply technology, to allow the production of hybrid pre-impregnated material of reduced thickness in an interyarn (tow-by-tow) architecture with controlled characteristics, such as the fiber ratios of the different tows, the thickness, the hybridization pattern and the fiber type.

However, fiber-by-fiber hybridization proved challenging and beyond the capacity of the existing industrial capabilities. As a result, a novel calendering process was introduced as an alternative for the production of hybrid thin-ply prepreg with a intrayarn: fiber-by-fiber architecture.

In parallel with the development of the fiber hybridization processes and the new hybrid thin-ply prepregs, the need for a reliable tool for microstructural analysis of hybrid thin-ply composites manufactured with dissimilar fibers (carbon-glass) or of the same type but with different grades was recognized. For that reason, microstructural sample preparation techniques were optimized to best reveal the microstructural features of thin-ply hybrid composite. A fiber identification and microstructural analysis tools was developed to capture and study the microstructural features of the new type of hybrid composites.

The microstructural analysis methods that were developed first ensured that the hybridization was achieved without any quality degradation and the new type of hybrid prepreg could achieve quality equivalence (homogeneous thickness, limited resin bleed and porosity, fiber alignment and packing) of that on non-hybrid thin-ply prepregs.

High-quality non-hybrid prepregs at low areal density were manufactured and the baseline properties of the selected materials were determined with an extensive experimental campaign. Several rolls of high-quality hybrid prepregs at low fiber areal density and with different levels of fiber dispersion (interyarn: tow-by-tow and intrayarn calendered fiber-by-fiber) were produced for a broad testing campaign conducted at EPFL. Multi-layer composite plates were manufactured with the produced hybrid thin-ply prepregs the morphology of which was examined with optical microscopy and the microstructural analysis techniques to ensure that the fiber hybridization was achieved without compromising the laminate quality. Overall, the quality of the composites was excellent, and the mechanical properties were as expected in tensile mode. It was also shown that the calendered prepregs were of high quality, although their degree of hybridization did not reach a level higher than with the same configuration, but simply autoclaved. This result indicates that the calendering process would need further optimisation, and that it may not be needed, at least for UD configurations, where the fibers already spread up to a few fiber diameters thin, spontaneously mingle under the pressure applied by the autoclave process.

Nevertheless, the goal of improving production efficiency and reducing costs remains a priority for the composites industry and is an ongoing area of research and development. This can be achieved with the new type of hybrid thin-ply prepreg materials that were developed. These materials simplify the composite manufacturing processes and broaden the hybrid design space with a new format of material that combines two different fibers in a single layer of prepreg tape of reduced thickness.

The work helps to bridge the gap between academic research and current industrial hybrid prepreg manufacturing capabilities, and help recognize that academic scenarios and model predictions may not always align with results that can be achieved in an industrial set up.

8.2 Suggestions for future implementations of the key finding of the study

The development of methods for creating thin-ply prepregs with different levels of hybridization has contributed to a more realistic understanding on the current benefits and limitations of prepreg manufacture within an industrial setting which is already one of the best in the field able to achieve such low levels of fiber areal weight.

By leveraging this collective knowledge and utilizing the unique expertise gained in this project, realistic solutions can be proposed to fine-tune the production of hybrid thin-ply prepregs. Additionally, there is a strong belief that to fully harness the available data for academic research, there should be a transition towards experimental fiber spreading and impregnation lines managed by academic institutions. This shift would allow comprehensive studies of various parameters without the limitations posed by proprietary methods. Moreover, it is anticipated that the successful fiber spreading, and hybridization achieved in this research can also be extended to thermoplastic matrix systems without the need for extreme modifications.

The use of ATL was also proposed as an automated method that can be used for achieving more complex hybrid architectures. For example, tow by tow hybrids could be manufactured using ATL deposition of two types of thin tapes, side by side. This would allow the use of non-hybrid thin ply tapes, and would lead to great design freedom to lay high strain fiber tapes exactly where needed in the part to provide higher translaminar toughness while preserving the high strength expected for thin ply configurations.

Finally, additional research is needed to fine-tune the microstructural analysis methods for hybrid composites, and to understand the relationship between the fiber arrangement and the resulting properties. This was the focus of other research work in the Hyfisyn project, however much is still to be done to be able to fully predict the quality and properties of these complex structures.

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Alexios Argyropoulos | Curriculum Vitae

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C.4. S. AhmadvashAghbash, G. Broggi, **A. Argyropoulos**, J. Cugnoni, V. Michaud, M. Mehdikhani and Yentl Swolfs. Translaminar Fracture Analysis in Hybrid Fibre-Reinforced Composites Through 4D In-Situ Synchrotron Tomography. ICCM23

C.5. G. Broggi, **A. Argyropoulos**, J. Cugnoni, and V. Michaud Translaminar Fracture Toughness Characterization in Fibre-Hybrid Composites: Effect of Hybridization. ICCM23

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