Circularity of Thermoplastic Matrix Composites and Applications

Nhlapo Thutswana Victor

Universidade do Minho
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Professor João Pedro Nunes

Doutor Paulo Antunes

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DIREITOS DE AUTOR E CONDIÇÕES DE UTILIZAÇÃO DO TRABALHO POR TERCEIROS

Licença concedida aos utilizadores deste trabalho

“Education is the most powerful weapon which you can use to change the world.”

-Nelson Rolihlahla Mandela

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All of you are a fundamental part of my story and I am deeply grateful for that.

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STATEMENT OF INTEGRITY

I hereby declare having conducted this academic work with integrity. I confirm that I have not used plagiarism or any form of undue use of information or falsification of results along the process leading to its elaboration.

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Universidade do Minho, Guimarães, 3 December 2023

Nhlapo Thutswana Victor

ABSTRACT

The concept of circularity in thermoplastic matrix composites refers to the ability to close the material loop by recycling and reusing these materials, thereby reducing waste and environmental impact. This thesis aims to be a relevant and new contribution to the study and analysis of the thermoplastic matrix composites' circularity and reuse. The demand for thermoplastic composites is continuously increasing because these materials offer many advantages over their thermoset counterparts, such as high toughness, long storage time, easy repair and recycling, and the ability to be thermoformed and heat welded. However, the manufacturing of thermoplastic composite parts using liquid moulding techniques (e.g., resin transfer moulding, vacuum-assisted resin infusion) is often impossible, and the melt processing, where high temperature and pressure are needed to impregnate the fibre reinforcement becomes also very complex and expensive due to the high viscosity of thermoplastics. These issues may be overcome using reactive processing where a fibrous preform is first impregnated by a low-viscosity mono or oligomeric precursor and the polymerization of the thermoplastic matrix then occurs in-situ. In this thesis, continuous fibre-reinforced composites were produced by using acrylic-based reactive thermoplastics as a matrix (e.g., polymethylmethacrylate (PMMA) such as Elium®), through VARI to obtain the composite plate.

An appropriate recycling route consisting of grinding composite plate, compounding through extrusion and compression moulding, was established to experimentally validate the mechanical performance of recycled TPCs. Characterisation was performed by flexural, DSC, TGA , optical microscopy testing and capillary rheometry. The experimental properties of the recycled material were found to be in-line with theoretical predictions and good mechanical properties that are recyclable and can be reused as secondary applications such as brackets .

The work carried out demonstrates that TPCs recycling is feasible and enables applications currently made by compounding CFRP with compatible thermoplastics. Conventional composites with thermoplastic matrix can be mechanically recycled obtaining materials with very good mechanical properties.

KEYWORDS: PMMA, VARI, Elium, Thermoplastic matrix, circularity, mechanical recycling, *in-situ* polymerization.

RESUMO

O conceito de circularidade em compósitos de matriz termoplástica refere-se à capacidade de fechar o ciclo do material reciclando e reutilizando estes materiais, reduzindo assim o desperdício e o impacto ambiental. Esta tese pretende ser um novo e relevante contributo para o estudo e análise da circularidade e reutilização dos compósitos de matriz termoplástica. Tem havido um aumento continuo da utilização de compósitos termoplásticos por eles oferecerem muitas vantagens relativamente aos seus homólogos termoendurecíveis, como sejam, maior tenacidade, período de armazenamento mais longo, maior facilidade de reparação e reciclagem e a possibilidade de serem termoformados e soldados usando calor. No entanto, a fabricação de peças em compósito termoplásticos usando técnicas de moldação por via líquida (por exemplo, a moldação por transferência ou a infusão de resina assistida a vácuo) é muitas vezes impossível e o processamento por fusão, com recurso a altas temperaturas e pressões para impregnar o reforço da fibra, torna-se também complexo e caro devido à alta viscosidade dos termoplásticos. Estes problemas podem-se superar usando o processamento reativo onde uma pré-forma fibrosa é primeiro impregnada por um precursor mono ou oligomérico de baixa viscosidade e a polimerização da matriz termoplástica ocorre então in situ. Nesta tese, produziram-se compósitos reforçados com fibras contínuas usando, como matriz, termoplásticos reativos à base de acrílico (por exemplo, polimetilmetacrilato (PMMA) como o Elium®), através de VARI para obter a placa composta.

Uma rota de reciclagem apropriada que consiste em moagem, composição por extrusão e moldagem por compressão, foi estabelecida para validar experimentalmente o desempenho mecânico dos TPCs reciclados. A caracterização foi realizada por testes de flexão, DSC, TGA, microscopia óptica e reometria capilar. As propriedades experimentais do material reciclado foram encontradas em linha com as previsões teóricas e boas propriedades mecânicas que são recicláveis e podem ser reutilizadas como aplicações secundárias, como barquetes.

O trabalho realizado demonstra que a reciclagem de TPCs é viável e viabiliza aplicações atualmente feitas através da combinação de CFRP com termoplásticos compatíveis. Compósitos convencionais com matriz termoplástica podem ser reciclados mecanicamente obtendo materiais com propriedades mecânicas muito bom.

PALAVRAS-CHAVE: PMMA, VARI, Elium, Matriz termoplástica, circularidade, reciclagem mecânica, polimerização *in situ*

CONTENT

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NOMENCLATURE

Acronyms

ABS	Acrylonitrile Butadiene Styrene
ASTM	American Society for Testing and Materials
BMC	Bulk moulding compound
C/PPS	Carbon fibre reinforced polyfenyleensulfide
CAGR	Compound annual growth rate
CFRP	Carbon fibre reinforced polymer
CM	Compression Moulding
DCM	Direct compression moulding
DSC	Differential Scanning Calorimetry
EoL	End-of-Life
FEA	Finite element analysis
FEM	Finite element method
FLD	Fibre length distribution
FRPC	Fibre reinforced polymer composite
FVD	Fibre volume distribution
FVF	Fibre volume fraction
GFRP	Glass fibre-reinforced polymer
GMT	Glass mat reinforced thermoplastic
ISO	International Organization for Standardization
LCA	Life-cycle-assessment
LCCA	Life-cycle-and-cost-assessment
LFT	Long fibre thermoplastic

MCM	Mixing compression moulding.
MMA	Methyl methacrylate monomer
PB	Polybutylene succinate
PBT	Polybutylene terephthalate
PEAK	(family of) Polyaryletherketone
PEEK	Polyetheretherketone
PEI	Polyetherimide
PM	Poorly mixed (material)
PMMA	Polymethylmethacrylate
PP	Polypropylene
PPS	Polyphenylene sulfide
RTM	Resin transfer moulding
SD	Standard deviation
SEM	Stereo electron beam microscope
SFT	Short fibre thermoplastic
TGA	Thermogravimetric Analysis
TPC	Thermoplastic composite
TPU	Thermoplastic polyurethane
TSC	Thermosetting composite
UD	Unidirectional (fibre orientation)
VARI	Vacuum Assisted Resin Infusion

1. INTRODUCTION

1.1. BACKGROUND AND MOTIVATION

The circularity of thermoplastic matrix composites refers to their ability to be recycled and reused, thereby reducing waste, and promoting sustainability. The plastics circular economy is a sustainable model where plastics remain in circulation longer and are reused and recycled at the end of their life span. To move toward eco-sustainable continuous fibre composites, one of the most effective solutions is to use thermoplastic matrix composites. These materials, in opposition to thermoset matrix ones, enable material circularity via mechanical or chemical recycling techniques. Composites based on thermoplastic matrices can be heated and remoulded without degradation, using conventional thermoplastic processing techniques such as injection moulding and compression moulding. In the context of the present thesis, liquid thermoplastic resins will be used to produce fibre-reinforced composite by traditional composite processing techniques and then, crush/grind the produced materials by using mechanical recycling techniques to obtain a new recycled product that could be processed by conventional thermoplastic moulding techniques.

Composite materials are on a rise. The material offers lightweight potential and can contribute to a reduction of emissions, especially in very defined areas of application such as the transportation sector. Within the continuous fibre composites' market, thermoplastic composites (TPC) account for an increasing market share in comparison to their thermosetting equivalent.

According to the market research Future, the Thermoplastic Composite Market Size based on consumption was at 6,841.5 thousand Tons in 2021, as shown in Figure *1* Thermoplastic composite current market. The Thermoplastic Composite market industry is projected to grow from 7,318.9 thousand Tons in 2022 to 13,297.8 thousand Tons by 2030, exhibiting a compound annual growth rate (CAGR) of 6.26% during the forecast period (2022 - 2030). These figures result from the increasing demand for lightweight products on account of rapid urbanization and increased consumer spending across the globe are driving the market growth [1].

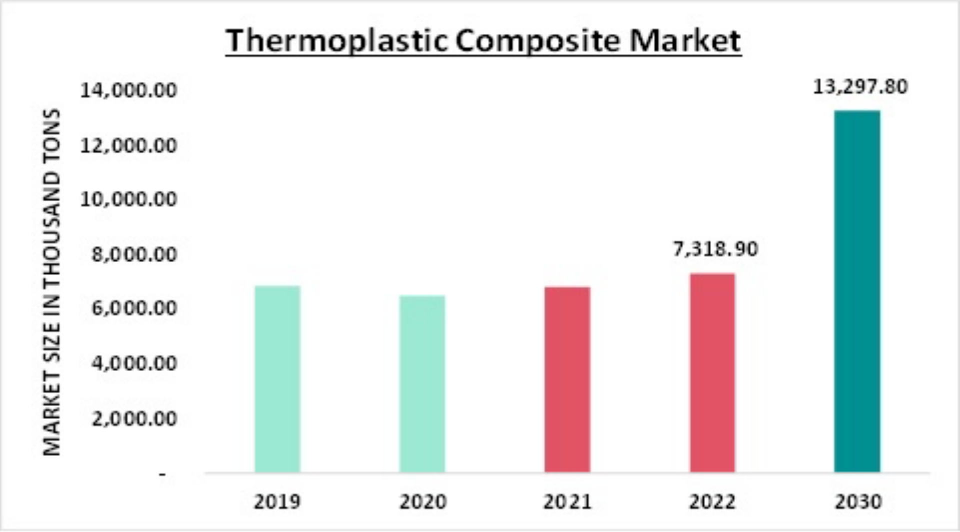


Figure 1 Thermoplastic composite current market adapted from ([1])

The automotive industry is increasingly focusing on lightweight products, which is expected to boost market growth globally. The demand for thermoplastic composite in the transportation industry is also on the rise, as it promotes low emissions of CO2 and ensures vehicles have higher efficiency and longer shelf life. Thermoplastic composites are used in various high-performance vehicle components like under-hood components, appearance grade components, roof components, door modules, front and end modules, and instrumental panels. This increasing focus on lightweight automotive products is anticipated to provide a boost to the demand for thermoplastic composites in the global market shortly.

Similarly, the adoption of thermoplastic composite in the aerospace industry is also growing. The new thermoplastic composites have several advantages like lower weight, indefinite shelf life, low moisture absorption, excellent thermal stability and chemical resistance, high toughness and damage tolerance, legendary solvent resistance, and relatively low dielectric constant. These qualities make thermoplastic composites ideal for the aerospace industry, and their high adoption is expected to fuel global demand for thermoplastic composite market products over the forecast period.[1].

1.2. PRESENTATION OF THE COMPANY

The Centre for Innovation in Polymer Engineering (PIEP) is a private association, with a technological and scientific matrix and a business management model. PIEP intends to provide a prompt response in the delivery of products and services, oriented to the R&D+i needs of companies in the plastics and related sectors, through innovation activities, technology transfer, technical-scientific consultancy, and services provision. In general, PIEP acts in terms of the provision of testing services and failure analysis, the development of new materials and products, processing technologies, and productive tools, based on the promotion of the principles of sustainable development (Circular Economy and Environment). PIEP also contributes to training and supporting the development of human resources with capacity and experience in industrial innovation in the field of polymer engineering.

1.3. OBJECTIVES AND SCOPE

The main objective of this thesis is to conduct comprehensive research into the circularity aspects of thermoplastic matrix composites and examine their potential applications across various industries. By analysing the recyclability, reusability, and renewability of these composites, we aim to evaluate their suitability for a more sustainable and environmentally friendly approach to material usage.

Objectives are :

- Produce recyclable composite parts using VARI using resin Elium.
- Mechanical recycling through grinding to obtain CFRP flakes.
- Compounding of ABS/CFRP through Extrusion.
- Compression Moulding of the homogenised dough.
- Mechanically characterise the recycled composite material through Flexural Tests.
- Physically characterise the recycled composite material using DSC (Differential Scanning Calorimetry) to quantify the glass temperature (Tg).
- TGA (Thermogravimetric Analysis) techniques will be conducted to quantify fibre volume content.

- Additionally, the dependence of the mechanical behaviour on temperature, fibre volume content, and fibre length will be exhaustively evaluated to enable a good characterization of the material.

This will enable the structural optimization of components produced by the recycled composite. To allow the production of geometrically more complex parts, a numerical rheological study on the recycled composite will be carried out to enable the optimization of the injection moulding process for this typology of recycled materials.

1.4. OUTLINE

To clarify and understand the contents exposed, the present dissertation was organized into five chapters.

Chapter 1 is the introduction. A framework of the project in question is carried out, a brief presentation of the company where it was developed is made, and the project motivations and objectives are stated.

Chapter 2 encompasses the state of the art of the topics covered by the project. It discusses the increasing interest in the sustainable and circular economy, which has led to a growing demand for sustainable materials like thermoplastic matrix composites. The concept of the circular economy and its relevance to thermoplastic matrix composites. It presents the principles and strategies of the circular economy, such as recycling, remanufacturing, and reuse, and how they can be applied to thermoplastic matrix composites to enhance their circularity. Overall, the state of the art provides a comprehensive understanding of the circularity aspects of thermoplastic matrix composites and their applications. It identifies research gaps and challenges and suggests potential future directions for advancing circularity in these materials.

Chapter 3 concerns the Methodology. In this chapter, the specific procedures and techniques employed to carry out the research study are outlined. Techniques such as tensile testing, flexural testing, impact testing, and thermal analysis techniques (e.g., DSC, TGA) used to obtain data on material behaviour and performance are presented and discussed.

Chapter 4 presents the results and discussion. As the name implies, this chapter serves the purpose of analysing the results of the experiments that were taken to build this thesis and giving a thorough discussion based on the results obtained.

Finally, chapter 5 sets out the conclusions reached, and a brief reference to future work that allows the continuation of this project. It emphasizes the importance of circularity in thermoplastic matrix composites for achieving sustainability goals and suggests potential strategies for enhancing circularity in these materials.

2. STATE OF ART

Thermoplastic matrix composites exhibit high circularity due to their recyclability and reusability properties. Unlike thermoset matrix composites, which undergo a chemical reaction during curing that irreversibly sets the material, thermoplastic matrix composites can be melted and reshaped multiple times without losing their mechanical properties [2]. This characteristic allows thermoplastic matrix composites to be recycled and reused, making them an attractive option for sustainable manufacturing processes. Moreover, the use of renewable and recyclable thermoplastic matrix materials in these composites further enhances their circularity. The ability of thermoplastic matrix composites to be recycled and reused has significant implications for their applications in various industries. For example, in the automotive industry, thermoplastic matrix composites can be used to produce lightweight components that not only reduce fuel consumption and emissions but also can be easily recycled at the end of their life cycle. Additionally, the circularity, excellent mechanical properties and low weight of thermoplastic matrix composites make them suitable for applications in industries such as construction and aerospace. In the construction industry, thermoplastic matrix composites can be utilized for structural elements and architectural designs that require both durability and sustainability [3]. Thermoplastic composites are often made up of multiple layers or different materials, such as fibres and resins. Separating these components can be challenging and requires specialized techniques, which can make the recycling process more complex and energy intensive. While certain thermoplastics can be recycled using relatively low-energy processes, the recycling of composites often involves high-temperature processes, such as thermal degradation or pyrolysis, which can be energy-intensive.

Thermosets and thermoplastics are both polymers, but they behave differently when exposed to heat. Thermoplastics always melt under heat while thermosets retain their form and stay solid under heat once cured and this makes thermoplastics ideal materials for being reprocessed and recycled. Thermosets, in contrast, can withstand higher temperatures without losing their shape, which makes them more durable but much more difficult to recycle and almost impossible to reprocess.

Thermoplastics are more aesthetically pleasing than thermoset polymers, yet thermosetting materials are still thought to be more aesthetically pleasing than other conventional materials, like metals. In-mould painting or coating is possible with these materials, including the direct application of coatings to the mould before the thermoset polymers are injected into it. Even in adverse weather, this method provides greater material adhesion and prevents chipping, cracking, or flaking.

2.1. THERMOPLASTIC-MATRIX COMPOSITES

Thermoplastic polymers consist of linear or branched polymer chains that form reversible polymer networks. When sufficient thermal energy is provided, thermoplastic polymers behave like a viscoelastic liquid and flow. When cooled, the material solidifies again. With relatively small degradation, the heating-cooling cycle can be carried out numerous times. This allows continuous processing, which quickly transforms thermoplastics into various products and applications, and enables reprocessing and recycling. However, when exposed to certain solvents (and heat), thermoplastic polymers dissolve completely [4]. As may be seen in Figure 2 there is a wide variety of thermoplastics used in composite materials with significantly different properties.

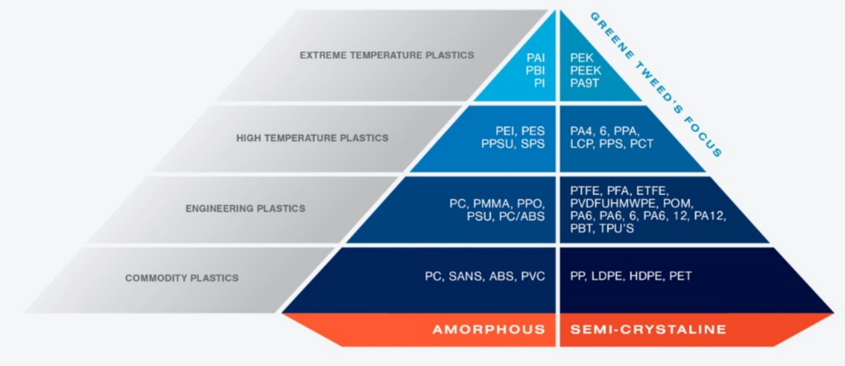
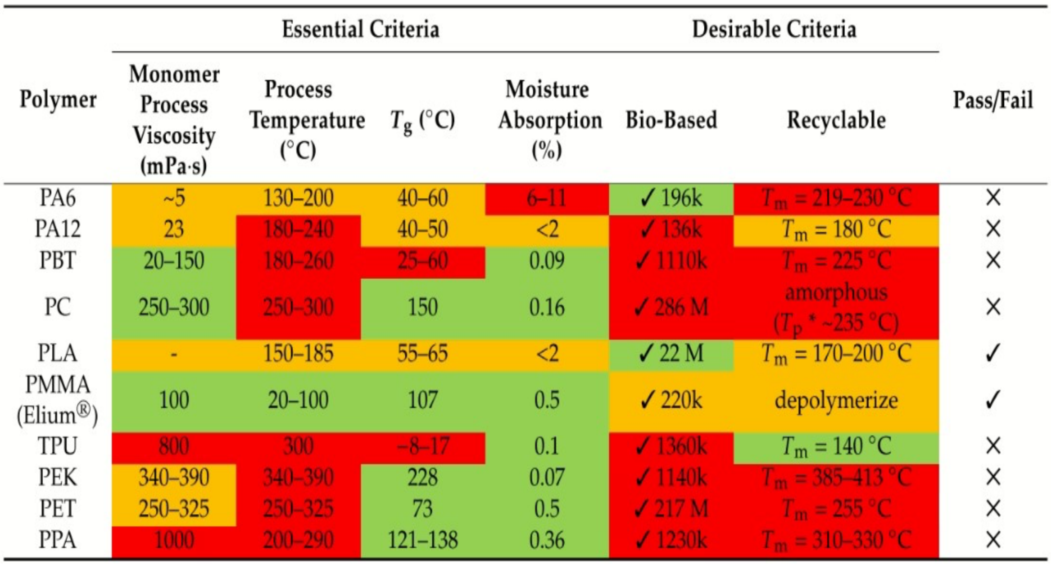


Figure 2 Thermoplastic materials (adapted from [4] *)*

According to the JEC Observer, thermoplastic composites represent currently 40–50% of polymer matrix composites, and this participation is constantly growing [5]. Short fibres are also the dominant reinforcement in thermoplastic composites (particularly, used in extrusion and injection production processes), while thermoset composites predominantly use continuous fibre reinforcements. Given the properties of thermoplastic matrices and the high viscosity that hinders the impregnation process, in continuous fibre thermoplastic composites, the reinforcement is pre-impregnated before the manufacture of parts, forming prepregs. These intermediate products are then used in the production of parts by processes that require pressure and temperature, which promote complete impregnation and interlaminar adhesion. The high temperature required for matrix melting is often a limitation to the use of fibrous natural fibre reinforcements because natural fibres (flax and hemp, thermally degrade between 170°C and 200°C) and make particularly difficult the production of large parts (wind blades, boats, etc.). The recent appearance of reactive thermoplastics looks to solve many of the problems associated with the processing of continuous fibre thermoplastic composites [5][6].

2.1.1. Reactive Processing of Acrylic-Based Thermoplastic Composites

In a reactive process of production of thermoplastic composites of continuous fibre, the impregnation of the dry reinforcement is carried out with a mono or oligomeric precursor that, at room temperature, is a low viscosity liquid and is subsequently polymerized in-situ in the mould, analogous to the moulding processes with liquid resin (e.g., RTM – Resin Transfer Moulding and VARI-Vacuum Assisted Resin Infusion) characteristic of thermosetting composites. In recent decades, several precursors have been developed for reactive thermoplastics of in situ polymerization, such as cyclic butylene terephthalate[7], caprolactam, caprolactam and L-lactido. Other relevant polymers, that can be used for reactive processing, are Polybutylene terephthalate (PBT), thermoplastic polyurethanes (TPU), Polyamide-6 (PA-6), and Polyamide-12 (PA-12). However, they have the significant disadvantage of requiring high processing temperatures [5][6][8]. Qin et al [8], in a review of the processing temperature and the corresponding viscosity of thermoplastic monomers commercially available for polymerization in situ, to produce composites reinforced with natural fibres, related the processing temperature with viscosity to different groups of polymers (see Figure 3), including thermoset. In general, thermoplastics require higher temperatures to achieve the viscosity required for liquid resin moulding processes (150°C for PA-6 and PA-12; 180°C for PBT; and 270°C for TPU). On the contrary, thermoset resins like epoxy, polyester, and vinyl ester only require ambient temperatures (see Figure 4) [5][6][8].



	Essential Criteria				Desirable Criteria		
Polymer	Monomer Process Viscosity (mPa·s)	Process Temperature (°C)	T_g (°C)	Moisture Absorption (%)	Bio-Based	Recyclable	Pass/Fail
PA6	~5	130–200	40–60	6–11	✓ 196k	T_m = 219–230 °C	×
PA12	23	180–240	40–50	<2	✓ 136k	T_m = 180 °C	×
PBT	20–150	180–260	25–60	0.09	✓ 1110k	T_m = 225 °C	×
PC	250–300	250–300	150	0.16	✓ 286 M	amorphous (T_p * ~235 °C)	×
PLA	-	150–185	55–65	<2	✓ 22 M	T_m = 170–200 °C	✓
PMMA (Elium®)	100	20–100	107	0.5	✓ 220k	depolymerize	✓
TPU	800	300	−8–17	0.1	✓ 1360k	T_m = 140 °C	×
PEK	340–390	340–390	228	0.07	✓ 1140k	T_m = 385–413 °C	×
PET	250–325	250–325	73	0.5	✓ 217 M	T_m = 255 °C	×
PPA	1000	200–290	121–138	0.36	✓ 1230k	T_m = 310–330 °C	×

Figure 3 Classification of different reactive thermoplastic monomers as to the potential in the product composite materials. (Adapted from [8]*))*

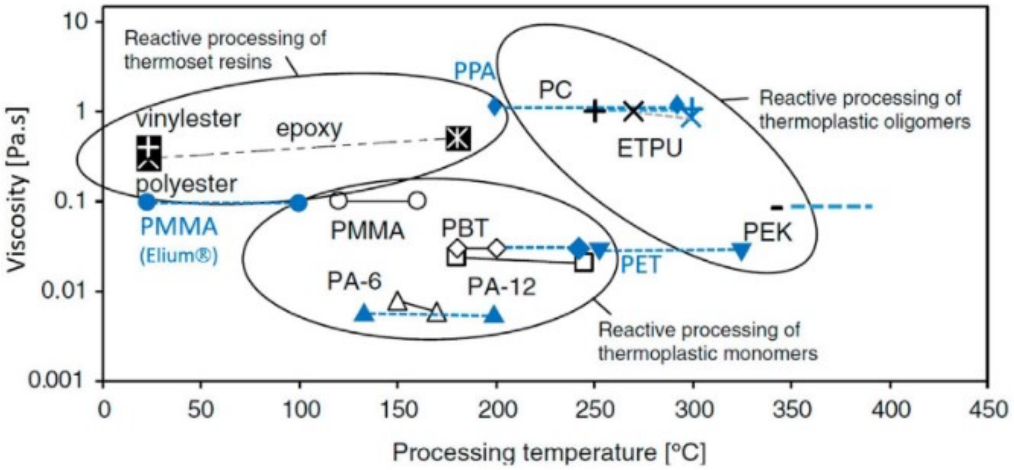
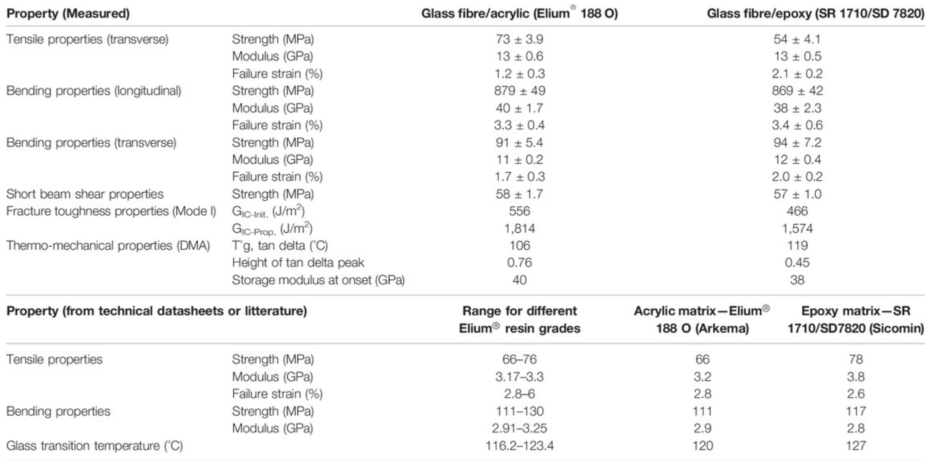


Figure 4 Processing temperature and viscosity ranges of various polymers suitable for reactive processing, compared with thermo-hardened resins (adapted from [5][6][8]*))*

The only material for *in situ polymerization at* room temperature available on the market is Arkema's Elium® thermoplastic resin [5][6]. Elium® liquid acrylic resin is a mixture of 2propythenic acid, 2-methyl-, methyl ester, or methyl methacrylate monomer (MMA) and acrylic copolymers. The combination of the resin with a compatible initiator, such as benzoyl peroxide, allows the conversion of MMA into its PMMA polymer, through diffusion-controlled reactions, in a free radical polymerization reaction. The polymerization reaction comprises three characteristic reactions [6].

Elium® reactive thermoplastic resin has a viscosity of approximately 100 MPa.s at 25°C as can be seen in Figure 5, being suitable for different moulding processes with liquid resin. In terms of mechanical properties Figure 5 has proved very similar to epoxy resins [5][6][8]. Figure 4 shows the results obtained for fibreglass laminates with epoxy matrix and Elium®. Obande et al [6] present a current literature review of the properties of reactive acrylic resin composite materials with fibrous reinforcement of carbon, glass, and flax.



Property (Measured)		Glass fibre/acrylic (Elium® 188 O)	Glass fibre/epoxy (SR 1710/SD 7820)
Tensile properties (transverse)	Strength (MPa)	73 ± 3.9	54 ± 4.1
	Modulus (GPa)	13 ± 0.6	13 ± 0.5
	Failure strain (%)	1.2 ± 0.3	2.1 ± 0.2
Bending properties (longitudinal)	Strength (MPa)	879 ± 49	869 ± 42
	Modulus (GPa)	40 ± 1.7	38 ± 2.3
	Failure strain (%)	3.3 ± 0.4	3.4 ± 0.6
Bending properties (transverse)	Strength (MPa)	91 ± 5.4	94 ± 7.2
	Modulus (GPa)	11 ± 0.2	12 ± 0.4
	Failure strain (%)	1.7 ± 0.3	2.0 ± 0.2
Short beam shear properties	Strength (MPa)	58 ± 1.7	57 ± 1.0
Fracture toughness properties (Mode I)	$G_{IC\text{-}Init.}$ (J/m^2)	556	466
	$G_{IC\text{-}Prop.}$ (J/m^2)	1,814	1,574
Thermo-mechanical properties (DMA)	T°g, tan delta (°C)	106	119
	Height of tan delta peak	0.76	0.45
	Storage modulus at onset (GPa)	40	38

Property (from technical datasheets or litterature)		Range for different Elium® resin grades	Acrylic matrix—Elium® 188 O (Arkema)	Epoxy matrix—SR 1710/SD7820 (Sicomin)
Tensile properties	Strength (MPa)	66–76	66	78
	Modulus (GPa)	3.17–3.3	3.2	3.8
	Failure strain (%)	2.8–6	2.8	2.6
Bending properties	Strength (MPa)	111–130	111	117
	Modulus (GPa)	2.91–3.25	2.9	2.8
Glass transition temperature (°C)		116.2–123.4	120	127

Figure 5 Comparison between glass fibre laminates with Acrylic Resin Elium® 188 O (Arkema) and epoxy resin SR 170/SD 7820 (Sicomin), produced by vacuum-assisted infusion: mechanical properties (adapted from [6]*)*

The reactive processing of acrylic-based thermoplastic composites involves the use of chemical reactions to enhance the properties and performance of the composites during their manufacturing process. This methodology can be applied to various thermoplastic matrix materials, such as poly(methyl methacrylate) (PMMA) and other acrylic-based resins, to improve their mechanical, thermal, and chemical properties. The following four different reactive processing techniques are being used to produce acrylic-based thermoplastic composites: i) reactive extrusion, ii) in-situ polymerization, iii) chemical modification and iv) surface treatment.

i. Reactive Extrusion:

Reactive extrusion is a widely used technique for processing acrylic-based thermoplastic composites. It involves the simultaneous melting, mixing, and reaction of the thermoplastic matrix material with reactive additives, such as crosslinking agents or chemical modifiers, in an extruder. The reactive additives react with the acrylic-based matrix resin during the extrusion process, leading to the formation of chemical crosslinks or modified polymer structures. This helps to enhance the mechanical strength, chemical resistance, and thermal stability of the composites.

ii. In Situ Polymerization:

In situ polymerization is another reactive processing technique used for acrylic-based thermoplastic composites. It involves the polymerization of monomers within the thermoplastic matrix material during the processing stage. Initiators or catalysts can be added to the acrylic-based resin, which then reacts and initiates polymerization reactions, resulting in the formation of a polymer network within the matrix. This in situ polymerization process can improve the strength, stiffness, and toughness of the composites by increasing the molecular weight and crosslink density of the matrix material.

iii. Chemical Modification:

Chemical modification techniques can be used to introduce functional groups or modify the chemical structure of the acrylic-based thermoplastic matrix material. This can be achieved through various chemical reactions, such as grafting, copolymerization, or crosslinking reactions. By selectively modifying the acrylic-based matrix resin, the properties of the composites can be tailored to specific applications. For example, the incorporation of polar or hydrophobic functional groups can improve the adhesion or compatibility with other materials or enhance the resistance to chemical agents.

iv. Surface Treatment:

Surface treatment techniques can be applied to modify the surface properties of acrylic-based thermoplastic composites. This can involve the use of plasma treatment, corona treatment, or chemical treatments to introduce functional groups or modify the surface energy of the composites. Surface treatment enhances the adhesion between the matrix resin and reinforcing materials, such as fibres or fillers, leading to improved mechanical properties and interfacial bonding in the composites.

The reactive processing of acrylic-based thermoplastic composites offers several advantages, such as the ability to tailor the properties of the composites, improve their performance, and enhance their processability. However, it requires careful control of reaction conditions, selection of appropriate reactive additives, and optimization of processing parameters to achieve the desired properties. The specific reactive processing technique used for acrylic-based thermoplastic composites will depend on the desired outcome and the compatibility of the reactive additives with the matrix resin [5].

2.1.2. Selecting possible routes for recycling

When it comes to recycling continuous fibre-reinforced thermoplastics, there are several possible routes to consider. The most suitable recycling route depends on factors such as the specific thermoplastic material used, the type and configuration of the fibre reinforcement, and the desired end-use application for the recycled material. Figure 6 shows three common recycling routes for continuous fibre-reinforced thermoplastics:

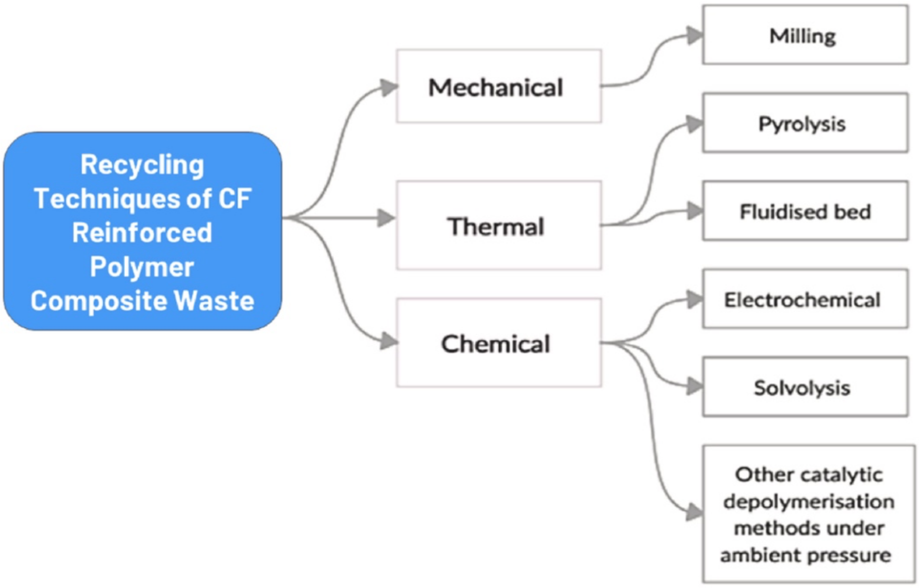


Figure 6 Classification of major recycling method adapted from [9].

2.1.2.1. Mechanical Recycling

Mechanical recycling of thermoplastic composites involves breaking down the composite material into small pieces or its original components, which can then be reused to manufacture new products. It involves shredding or grinding the thermoplastic composite material into smaller pieces, if needed separating the fibres from the matrix, and then reusing the little pieces obtained or the separated components[10]. This method is particularly suitable for thermoplastics with relatively low melting points, such as polypropylene (PP) or polyethene (PE). The separated fibres can be reused for reinforcement in new thermoplastic composites, while the matrix can be melted and processed into new thermoplastic parts or pellets. The mechanical recycling process may vary depending on the specific type of thermoplastic composite being recycled and the desired end application. Additionally, factors such as the quality and properties of the recycled material can be affected by the original composite composition, processing conditions, and the number of recycling cycles it has undergone.

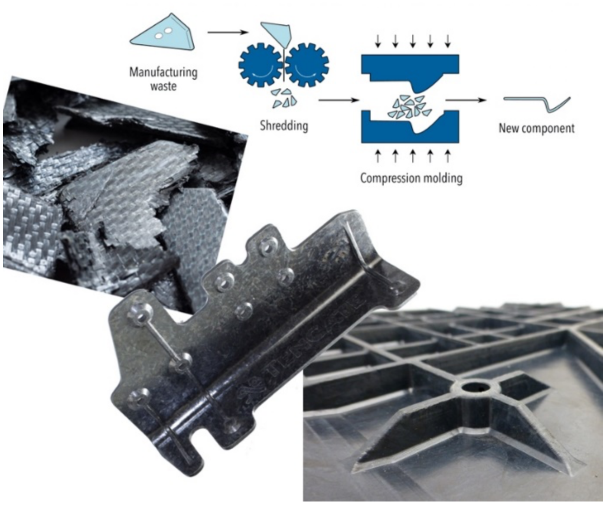


Figure 7 Mechanical recycling route for thermoplastic composites

2.1.2.2. Chemical Recycling

Chemical recycling involves using various chemical processes to break down the thermoplastic matrix and recover the fibres. This method applies to thermoplastics with higher melting points, such as polyamide (PA) or Polyethylene terephthalate (PET). The matrix material is typically dissolved or depolymerized, allowing the fibres to be separated and recovered. The recovered fibres can then be used for composites.

2.1.2.3. Pyrolysis

Pyrolysis is a thermal recycling process that involves subjecting the thermoplastic composites to high temperatures in an oxygen-deprived environment. This causes the materials to decompose into their constituent components, including the fibres and the matrix. The recovered materials can be further processed and used in various applications. Pyrolysis is suitable for thermoplastics with high thermal stability, such as polyphenylene sulphide (PPS) or polyetherimide (PEI).

2.1.2.4. Recycling of TPCs by low shearing mixing

Based on the study of previous literature on the recycling of TPCs, a recycling solution was implemented in a research project in collaboration with Thermoplastic Composites Research Centre, Thermoplastic Composites Application Centre, GKN Fokker, Toray Advanced Composites, Cato Composite Innovations, Dutch Thermoplastic Components, and Nido

Recycling Techniek. The processing steps start from A to E, as shown in Figure 8, which also contains photographs of the material at each processing step. The recycling route starts with collecting post-industrial scrap from various manufacturing sites and transporting it to a recycling site. The collected TPC scrap is then crushed using multi-shaft crushers to flake a few centimetres in size. A sieving step is added, if necessary, to recover only flakes of the desired size. Remanufacturing comprises two stages. The flakes are first fed into a low-shear mixer along with polymer granules to melt the polymer fraction and mix the flakes and polymer. The addition of polymer granules, virgin in this study, decreases the fibre fraction of recycled material. The blender used was selected for its ability to blend multi-ply flakes made with fabric into matted bundles, preventing fibre breakage. A molten and mixed mass is extruded from the mixer and immediately transferred to a press for compression moulding. The moulding phase takes place in an isothermal mould at a temperature below the melting point of the polymer. Thus, a waiting time of just a few minutes is enough for the part to cool down and be released. The reduced fibre fraction is essential for this step so that the mass can flow into the mould cavity [11]. The maximum fibre content that limits the material flux into the mould cavity depends on several factors including the type and viscosity of the resin, the size and shape of the reinforcing fibres, and the geometry of the mould cavity.

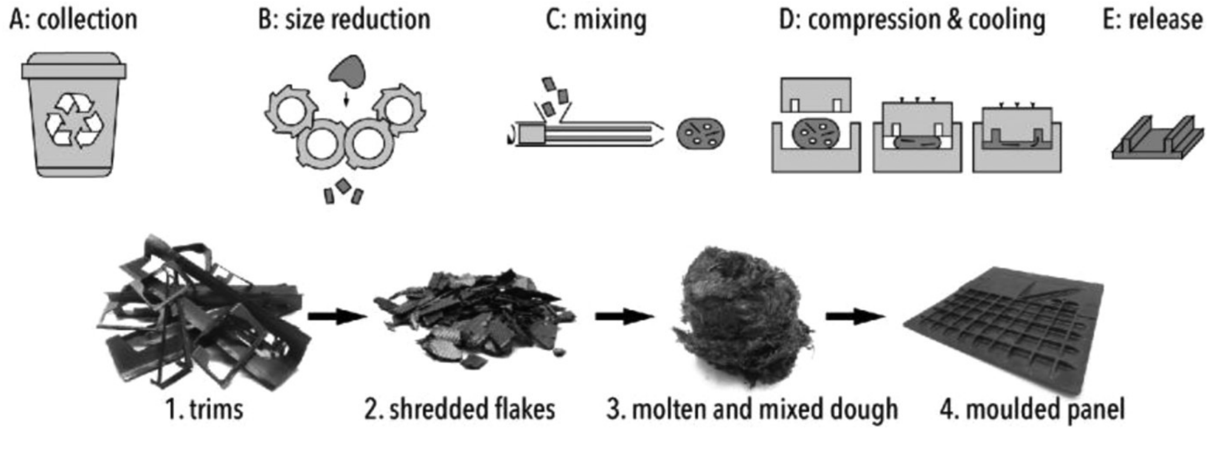


Figure 8 Flow chart of low shear mixing recycling of TPC taken from ([11]*).*

2.1.3. Composites Recyclability

The growing concern with the effects caused by human action in the biosphere, the legal requirements for recycling, and the costs imposed by it, require reformulating the life cycle of composite materials, from their origin to the end of life, with the main objective of revaluation and reuse, to minimize the amount of waste produced at the end of the product cycle.

In new products, the application of eco-design concepts allows an approach to try to solve the problems, focusing the approach on the materials to be used, on the design of the component, its maintenance and repair, and targeting, the increase in lifetime and an end of life thought of reuse, disposal, and recycling. Reuse may provide for the incorporation of mechanically treated material into new products, or the application or use of part of the parts in new applications and new products [10]. The adoption of "green composites", incorporating fibres and matrices of natural origin, also creates concerns about the resources to obtain these raw materials [11].

Composite materials consist of two or more materials in different phases, which are heterogeneous mixture, a reinforcing material and a matrix. The reinforcing material, usually in the form of fibres (glass, carbon, natural, aramid, or another type), which attribute stiffness and resistance to composite; and the matrix, in the polymeric, thermoplastic, or thermoset, which functions as an element of union and consolidation, allowing the shape of the piece, and transmitting the loads to the fibres.

Thermoset matrix, once polymerized, are not re-processable. In turn, thermoplastic matrices allow heat to be melted or remodelled when heat is supplied, allowing easy recycling, although some limitations apply [12]. Figure 9 shows the effects of material non-circularity when thermoset matrixes are considered to produce components. In the example shown, wind blades are piled up in a landfill.

Figure 9 - pilling up of wind blades in a landfill taken from [12].

The use of recyclable materials such as thermoplastic matrix during the production of composite materials is one of the best solutions to deal with waste treatment of composites. When heated above their melting temperature, thermoplastics melt and may easily processed into new shapes, which means they can be re-used directly into new products [11]. However, it is very important to note that recycling may cause a reduction of some of the mechanical properties of the material. Tri et al. [12] studied composites made from recycled polypropylene PP reinforced with natural fibres (bamboo fibres). They have shown that thermoplastic matrices such as polyethylene (PE) [polypropylene (PP)], and polystyrene (PS) are the most used due to their satisfactory treatment temperature. The required temperature for producing composites with these thermoplastics is less than 220°C, which makes it possible to avoid thermal deterioration of the lignocellulosic fibres. In addition to these synthetic resins, bio-based and biodegradable thermoplastics are appearing such as polylactide and poly (butylene succinate) (PBS) These materials present the best solution to face current environmental problems related to plastic pollution. However, despite good environmental performance, their characteristics (mechanical, thermal, etc.) strongly limit their use for structural applications.

By using thermoplastic resin as a matrix and natural fibres, recyclable materials with less environmental impact can be produced. But an important question must be asked What process can be used to recycle this type of material? Recycling thermoplastic composites can be accomplished using several methods such as mechanical processes (mainly grinding), pyrolysis and other thermal processes, and solvolysis. A comprehensive overview of the technologies for recycling composite materials was given by Henshaw et al. [13]. Moreover, Pimenta et al. [14] present in their study an excellent technology review on recycling carbon fibre-reinforced polymers for structural applications. Also, recycling technologies have been addressed in the handbook published by Goodship [15]. This study focuses specifically on a recycling method based on a thermomechanical process. This technique, which will be presented later, is inspired by the progress achieved in recent years in the processes of implementing cut thermoplastic prepregs. It consists of reshaping the waste into the required component by the combined action of temperature and pressure. The material manufactured after this recycling process is a short-fibre-reinforced composite because the raw continuous fibre material was previously cut into small prepreg composites before the final compression process. Consequently, this recycling method causes loss of performance in the composites due to the high reduction of fibre length and inability to control fibre orientation, not also mention the polymer degradation and deterioration of the fibre-matrix interface. In addition, it should be noted that the thermocompression cycle depends on three parameters: pressure, holding time, and temperature. In the literature, many studies are interested in the influence of such parameters on composite quality. As the melting temperature of several thermoplastics is too high is imperative to use a short manufacturing time to avoid damage to the natural fibres.

2.2. CARBON FIBRES

Carbon fibre-reinforced polymer matrix composite (CFRP) materials are increasingly used in a wide range of industries, including air, land, and sea vehicles, wind turbines, storage tanks, and sports equipment. CFRP composites have seen significant advancements and wide-ranging applications in various industries. Their properties, such as lightweight, high strength, and chemical stability, make them highly desirable for use in aerospace, automobile, military, sports sector, and civil engineering structures. One of the key advantages of CFRP composites is their high specific stiffness and strength. These properties make CFRP composites ideal for applications where weight reduction and enhanced performance are crucial in almost all advanced structural applications [13]. CFRP composites consist of high-strength carbon fibres embedded in a polymer matrix, with epoxy resin being commonly used as the matrix material due to its good mechanical properties [14]. The use of woven CFRP composites further enhances their mechanical properties and makes them suitable for applications that require balanced in-plane properties, excellent durability, and enhanced resistance to impact damage[13]. Furthermore, the aligned carbon fibres and cohesive matrix in CFRP composites contribute to their superior specific strength and stiffness, enabling them to withstand high loads and provide structural integrity in demanding environments. Overall, the advancements and applications of carbon fibre-reinforced polymer matrix composites have revolutionized several industries.

2.2.1. Global market for carbon fibre composites

The global market of carbon fibre composites, specifically carbon fibre-reinforced polymer matrix composites, has witnessed significant growth in recent years. These lightweight and high-strength materials have found extensive applications in various industries such as aerospace, automotive, wind energy, and sports equipment. The global carbon fibre market which was valued at USD 6.5 billion in 2022 is projected to reach USD 21.7 billion by 2032, growing at a cagr 12.9% from 2022 to 2032. Figure 10 shows the global demand and market growth which is attributed to the carbon fibres' excellent mechanical properties, superior chemical resistance and temperature tolerance, low weight, and distinguished thermal expansion.

Furthermore, the imposition of stringent eco-friendly regulations in developed and developing countries is projected to offer abundant growth opportunities to the CFRP market in the upcoming years. However, the high cost of carbon fibre still is a fundamental concern associated with carbon fibre market growth. Carbon fibre composites, including carbon fibre tapes, are only employed in luxury vehicles in the automobile industry and the aerospace and defence industry accounted for a share of 44.2% in terms of value in the carbon fibre market in 2022 and is projected to reach USD 8.5 Billion by 2032 at a CAGR of 11.6%[1], [15].

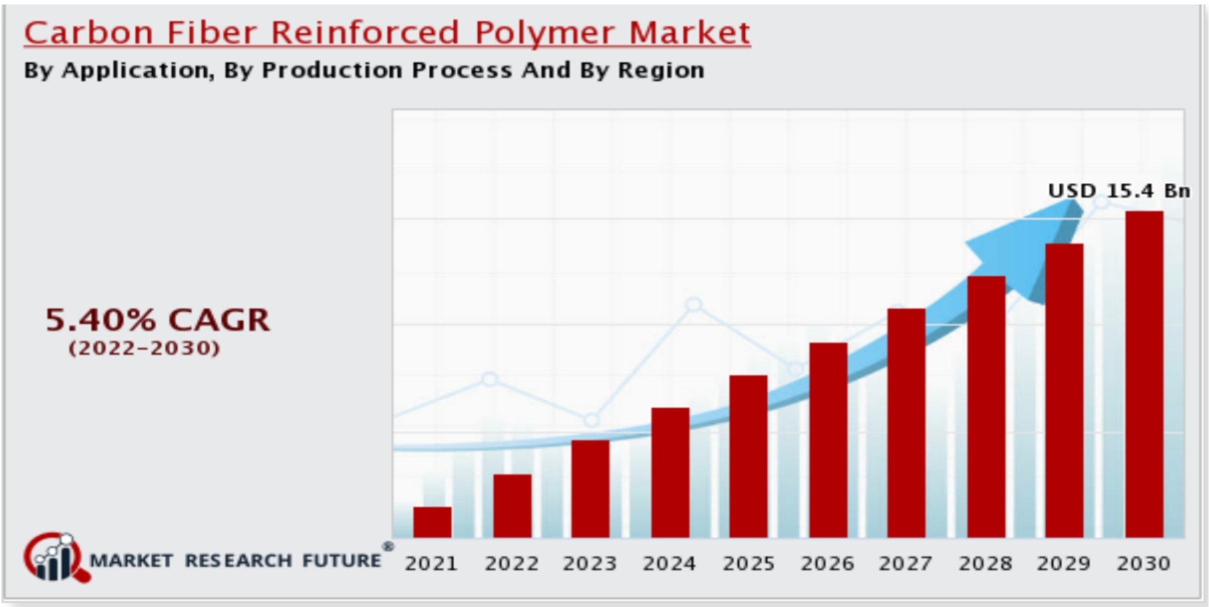


Figure 10 Global market for Carbon fibre taken from ([12]*).*

2.2.2. The Benefits and Properties of Carbon Fibre Reinforced Polymer Matrix Composites

Table 1 Advantages of the use of CFRP Composites

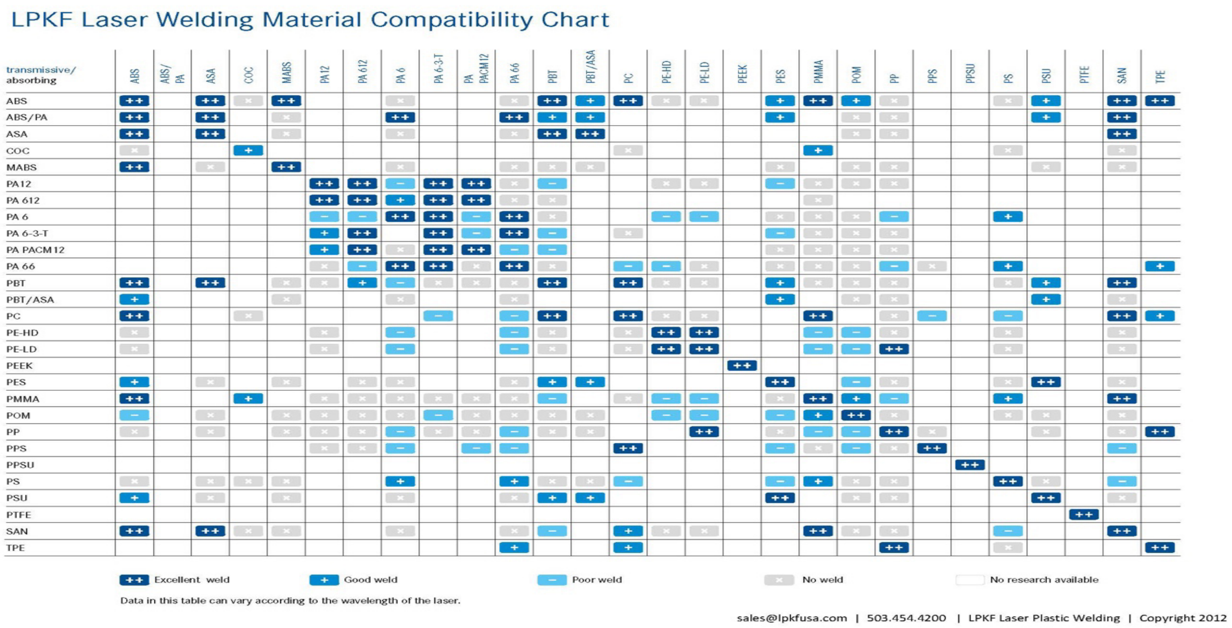
Properties	Benefits
High strength-to-weight ratio	CFRP composites are incredibly lightweight while possessing high strength and stiffness. These materials offer a strength-to-weight ratio that is much higher compared to traditional materials like steel and aluminium. This allows to produce lighter and more fuel-efficient vehicles, aircraft, and structures.
Excellent stiffness	CFRP composites have a high elastic modulus, which means they are extremely rigid and do not deform easily under stress. This makes them suitable for applications where stiffness is crucial, such as in structural aerospace, defence, automobile and sporting goods applications.
Fatigue resistance	CFRP composites have excellent fatigue resistance, making them ideal for applications subjected to cyclic loading and vibrations. This property allows for long-term durability and reliability, especially in structures and components under constant stress.
Thermal stability	CFRP composites can withstand high-temperature environments (especially in the absence of oxygen) and present very low shrinkage (carbon fibres have negative linear expansion coefficient). These properties are very important for many applications in aerospace, automotive, and other industrial markets.
Design flexibility	CFRP composites offer design freedom due to their ability to be moulded into complex shapes and geometries. This allows manufacturers to create innovative and optimized products with enhanced performance and functionality.
Electrical conductivity	As carbon fibres are a conductive material, CFRP composites can be engineered to have specific electrical conductivity properties, making them suitable for applications in the electronics and electrical industries.
Reduced costs	Although CFRP composites can initially be more expensive than traditional ones, their lightweight nature can lead to significant cost savings in transportation and installation. Additionally, CFRP composites require minimal maintenance, leading to long-term cost savings.

2.2.3. COMPATIBILITY

ABS (Acrylonitrile Butadiene Styrene) is compatible with Carbon Fibre (CF) reinforced with Elium 188XO. Elium 188XO is a thermoplastic resin created by Arkema which is specifically designed for carbon fibre reinforcement. Elium 188XO has excellent adhesion properties to carbon fibres, allowing for strong bonding between the resin and the fibres. This results in improved performance and mechanical properties in the final composite material.

ABS is a widely used thermoplastic polymer known for its good impact resistance, toughness, and low cost. It can be easily processed through techniques such as injection moulding or extrusion. When ABS is combined with CF reinforced with Elium 188XO, the resulting composite material benefits from both the strength of carbon fibres and the impact resistance of ABS. This combination can lead to a higher strength-to-weight ratio compared to ABS alone.

However, it is important to note that the compatibility of ABS with CF reinforced with Elium 188XO may still depend on various factors such as processing conditions, fibre content, and the specific requirements of the application. Therefore, it is advisable to conduct compatibility tests. Figure 11 shows how PMMA is compatible with ABS, and they have excellent weld. In addition, reaching out to the manufacturers or suppliers of both ABS and Elium 188XO can provide valuable insight and guidance regarding their compatibility and any precautions or recommendations for combining them.



LPKF Laser Welding Material Compatibility Chart

transmissive/ absorbing	ABS	ABS/ PA	ASA	COC	MABS	PA 12	PA 612	PA 6	PA 6-3-T	PA PACM12	PA 66	PBT	PBT/ASA	PC	PE-HD	PE-LD	PEEK	PES	PMMA	POM	PP	PPS	PPSU	PS	PSU	PTFE	SAN	TPE
ABS	++		++	×	++			×			×	++	+	++	×	×		+	++	+	×			×	+		++	++
ABS/PA	++		++		×			++			++	+	+					+		×	×				+		++	
ASA	++		++		×			×			×	++	++							×	×						++	
COC	×			+										×					+					×			×	
MABS	++		×		++			×			×	×	×					×		×	×				×		×	
PA12						++	++	−	++	++	×	−			×	×		−	×	×	×							
PA 612						++	++	+	++	++	×	×							×									
PA 6						−	−	++	++	−	++	×			−	−		×	×	×	−			+				
PA 6-3-T						+	++		++	−	++	−		×				−	×	×	×							
PA PACM12						+	++	×	++	++	−	−						×	×	×	×							
PA 66						×	−	++	++	×	++	×		−	−	×		×	×	×	−	×		+				+
PBT	++		++		×	×	+	−	×	×	×	++		++	×	×		+	×	×	×			×	+		++	
PBT/ASA	+				×			×			×							+		×	×				+		×	
PC	++			×					−		−	++		++	×	×			++		×	−		−			++	+
PE-HD	×					×		−			−	×		×	++	++			−	−	×			×			×	
PE-LD	×					×		−			−	×		×	++	++			−	−	++			×			×	
PEEK																	++											
PES	+		×		×		×	×			×	+	+					++		−	×			×	++		×	
PMMA	++			+		×	×	×	×	×	×	−		×	−	−		×	++	+	−			+			++	
POM	−		×		×	×	×	×	−	×	×	×	×		−	−		−	+	++	×			×	×		×	
PP	×		×		×	×	×	−	×	×	−	×	×			++		×	−	−	++	×			×		×	++
PPS						×	×	−		−	−			++				−	×	−	×	++					−	
PPSU																							++					
PS	×		×	×	×			+			+	×	×	−				−	+	×	×			++	×		−	
PSU	+		×		×			×			×	+	+					++		×	×				++		×	
PTFE																										++		
SAN	++		++	×	×			×			×	−		+	×	×			++	×	×			−			++	
TPE											+			+							++			×				++

++ Excellent weld
\+ Good weld
− Poor weld
× No weld
☐ No research available

Data in this table can vary according to the wavelength of the laser.

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Figure 11 Compatibility chart of thermoplastics polymers

2.3. PROCESSING TECHNIQUES

Thermoplastic composite materials have gained significant attention in various industries due to their unique properties and processing techniques. These materials offer a wide range of advantages, including a high strength-to-weight ratio, excellent chemical resistance, and superior impact resistance.

2.3.1. Processing continuous fibre thermoplastic composites by Vacuum Infusion

Vacuum infusion is a technique used to produce high-performance continuous fibre-reinforced composite materials. This process involves impregnating continuous fibres with a thermoplastic or thermosetting resin using a vacuum system. The advantage of using Vacuum Infusion for processing continuous fibre thermoplastic composites is that it allows for precise control over resin impregnation, resulting in improved mechanical properties and enhanced overall performance of the composite material. The process is particularly suitable for type thermoplastic composites that are based on reactive monomers because it overcomes the challenge of the high viscosity of molten thermoplastics which cannot be infused through the stack of fabric using conventional composite moulding techniques.

In vacuum infusion, the liquid resin is drawn into the fibres through the application of negative pressure (vacuum), ensuring complete fibre wetting and void elimination, In well-executed vacuum infusion processes, typical void contents range between 1% and 5%. Achieving void contents below 1% is challenging but feasible with careful attention to detail during the infusion process, such as effective degassing methods, optimizing resin flow, and ensuring thorough wetting of the fibres.

The method also allows for the potential of fast in situ out-of-autoclave processing, reducing work time and costs while replacing traditional materials like metals and wood in various applications. Thermoplastic composites offer several advantages, including the potential for fast in situ out-of-autoclave processing and automation, which can significantly decrease work time and costs [16].To overcome the high viscosity of the molten thermoplastic matrix and achieve optimal impregnation of the fibre, a variety of manufacturing processes have been developed, with vacuum Infusion being one of the most preferred techniques for processing continuous fibre thermoplastic composites when liquid thermoplastic resins capable of being processed at temperatures close to that of ambient are used [17].

2.3.2. Processing discontinuous fibre thermoplastic composites

Processing discontinuous fibre thermoplastic composites presents certain challenges due to their characteristics. One of the main challenges is achieving proper impregnation of the fibre bundle due to the high viscosity of the molten matrix. To overcome this challenge, innovative processing strategies have been developed, such as in-situ polymerization of an acrylic liquid thermoplastic resin after fibre impregnation [18]

This approach allows for better control of the viscosity and ensures effective impregnation of the fibres. Additionally, the use of prepregs is commonly preferred in industrial practice for manufacturing thermoplastic composites as they help overcome the problem of low impregnation [17]. By using pre-impregnated reinforcing yarns or tows, the viscosity of the molten matrix can be better controlled, and the fibre impregnation can be improved. These strategies enable the production of high-quality thermoplastic composites with good fibre distribution and strong bonding between the fibres and matrix. One of the main challenges in processing discontinuous fibre thermoplastic composites is achieving proper impregnation due to the high viscosity of the molten matrix. To address this challenge, innovative processing strategies have been developed, such as in-situ polymerization of an acrylic liquid thermoplastic resin after fibre impregnation [18]. This approach allows for better control of the viscosity and ensures effective impregnation of the fibres.

Processing discontinuous fibre thermoplastic composites involves several steps, including material preparation, mixing, preheating, moulding, cooling, finishing and post-processing. This step-by-step process is described below:

i. Material selection: Choose the appropriate thermoplastic matrix material and reinforcing fibres. Common thermoplastics used include polypropylene (PP), polyamide (PA), and polyethene (PE).

ii. Material preparation: Cut or chop the reinforcing fibres (such as glass, carbon, or aramid) into a predetermined length. The fibre length typically ranges from a few millimetres to a few centimetres.

iii. Mixing: Combine in a mixing chamber the thermoplastic matrix with the chopped fibres. The fibres should be evenly dispersed in the matrix to ensure uniform reinforcement. This can be achieved by using mechanical mixing or melt compounding techniques.

iv. Preheating: Preheat the mixture to a specific temperature above the melting point of the thermoplastic matrix, but below the degradation temperature of the matrix and fibres. This helps to soften the matrix and improve its flow during moulding.

v. Moulding: Transfer the preheated mixture into a mould or die cavity, which has the desired shape of the final composite part. Apply pressure to ensure proper consolidation and fibre impregnation.

vi. Cooling: Allow the moulded part to cool and solidify, leading to the formation of a solid composite structure. Cooling can be achieved through natural air cooling or by using cooling systems if a faster cycle time is required.

vii. Trimming and Finishing: After cooling, remove the part from the mould and trim any excess material. Further finishing processes like surface treatment, painting, or coating can be applied as needed.

viii. Post-processing: Depending on the desired properties and application requirements, additional post-processing steps such as annealing, welding, or machining may be performed.

Based on the research it is important to note that the specific processing parameters (e.g., temperature, pressure, cooling rate) depend on the chosen materials, part geometry, and desired performance characteristics. Therefore, optimization and validation of the process parameters are essential to achieve the desired quality and mechanical properties of the resulting composite parts.

2.3.3. Direct Compression Moulding

Compression moulding is a widely used manufacturing process for producing thermosets, thermoplastics, elastomers, and natural rubbers. This process is particularly useful for producing dimensionally precise, high-strength, temperature-resistant parts with good surface quality in high volumes. Parts can be produced in various sizes, thicknesses, and complexities at a better cost.

Direct Compression Moulding of thermoplastic composites is a manufacturing process used to create strong and lightweight components. It involves the compression of layers of thermoplastic composite material, typically consisting of a matrix material reinforced with fibres or another strengthening agent. The direct compression moulding process involves the following basic steps:

1. Preparing the composite material: The thermoplastic matrix material and reinforcing fibres are selected and prepared according to the desired properties of the final component.

2. Layer stacking: The composite material is layered in the desired orientation and structure to achieve the final required properties. The layers are typically arranged with alternating fibre orientations to maximize strength and stiffness.

3. Mould preparation: A mould is prepared based on the desired shape and dimensions of the final component. The mould may consist of two halves or multiple sections depending on the complexity of the part.

4. Heating and compression: The layered composite material is placed between the mould halves, and after heating pressure is applied. The heat melts the thermoplastic matrix material, allowing it to flow and fill the gaps between the reinforcing fibres. The pressure helps to consolidate the layers and remove any voids or air pockets.

5. Cooling and solidification: After the required time under heat and pressure, the mould is cooled to allow the thermoplastic matrix material to solidify and harden. This ensures the component retains its shape and provides the desired mechanical properties.

6. Demoulding and finishing: Once the component has sufficiently cooled and solidified, it is carefully removed from the mould. Any excess material or flash is trimmed or finished to achieve the desired final shape and appearance.

Production processes for thin thermoplastic composites that are subjected to direct compression moulding (DCM) have been examined in several studies. Rasheed et al. [19] investigated the recycling of chopped woven flakes of semi-preg C/PPS arising from nesting debris, whereas most of this research considers virgin material. The material is inserted immediately into a mould at the beginning of the procedure described in this research, which comprises various steps that are represented. After that, the mould is heated above the melting point of the material. To maintain contact, the material is held inside the heated mould under a small amount of pressure until the required temperature is reached. A predetermined moulding pressure is applied to the material for a predetermined dwell period once the material reaches the desired temperature. Finally, the mould and material are cooled with constant moulding pressure before the final part is released.

This recycling process for thermoplastic composites is relatively simple and like the process used for Bulk moulding compounds (BMC). Results showed a manufacturing route providing design freedom not feasible with continuous fibres: introduction of design features including ribs, thickness variations, and bosses, offers function integration and geometrically optimized structures.

According to the study of Thomas Alwart [20] a wide range of strength values may result from stress concentrations around the edges of the flake. Thin, flexible prepreg flakes were employed in this study. Parts having large fibre volume fractions (>50% FVF) that have a comparatively high rigidity might be moulded. On the other hand, parts made by DCM showed low levels of homogeneity; voids, pit holes, jams, and resin ridge areas were common. The flakes' meso-structure may limit their qualities by resulting in a poor load transfer load. Additionally, problems might arise during manufacturing, such as jamming, which may cause damage to the mould. However, because there is no mixing phase involved, adding polymer to lower the fibre content—for example, to make processing easier—can result in uneven and localised fibre contents.

2.3.4. Mixing Compression moulding

Utilizing the right techniques is crucial for achieving optimal results when processing thermoplastic composites. One such technique that plays a significant role in this process is mixing compression moulding. This technique involves the careful blending of thermoplastic materials and reinforcing fibres to create a composite material with enhanced mechanical properties. Mixing compression moulding allows for the uniform distribution of fibres throughout the matrix, resulting in improved strength, stiffness, and durability of the composite material. The technique ensures that the fibres are evenly dispersed, which reduces the likelihood of weak spots and improves the overall quality of the final product. With the uniform distribution of fibres throughout the matrix, the composite material can withstand higher loads and stresses, making it ideal for use in high-performance applications.

The process begins with the selection of suitable thermoplastic resins and reinforcing fibres based on the desired end-product requirements. These materials are then combined using specialized mixing equipment to ensure a homogeneous blend. The mixture is then subjected to compression moulding, where heat and pressure are applied to shape it into the desired form[21].

One of the key advantages of mixing compression moulding is its ability to produce complex shapes with high precision. This makes it an ideal choice for industries such as automotive, aerospace, and consumer goods where intricate designs are often required.

Furthermore, this technique offers excellent repeatability and scalability, allowing manufacturers to efficiently produce large quantities of thermoplastic composite parts without compromising on quality.

In conclusion, mixing compression moulding is a vital processing technique in the realm of thermoplastic composites. Its ability to create strong and durable composite materials with intricate designs makes it an invaluable tool for various industries. By leveraging this technique effectively, manufacturers can unlock new possibilities in product development and drive innovation in their respective fields.

2.3.5. Extrusion of thermoplastic composites

Extrusion is a widely used production technique for thermoplastic composites [22]. It involves forcing the molten thermoplastic material through a die to create a continuous shape or profile. This process allows for precise control over the shape and dimensions of the final product and is suitable for both simple and complex geometries. The extrusion process for thermoplastic composites offers efficient and versatile manufacturing capabilities. Additionally, extrusion allows for the incorporation of reinforcing fibres or fillers into the thermoplastic matrix, resulting in enhanced mechanical properties and improved performance of the composite material. Extrusion is a crucial method for the large-scale production of thermoplastic composites. Thermoplastic composites can be extruded using various methods, allowing for precise control over shape and dimensions [23]

These methods include single-screw extrusion, twin-screw extrusion, and coextrusion. By using single screw extrusion, thermoplastic composites can be processed into continuous profiles with a homogeneous mixture of the matrix and reinforcements. Twin screw extrusion, on the other hand, offers the advantage of better mixing and dispersion of reinforcement materials, resulting in improved mechanical properties of the final thermoplastic composite product. The extrusion process for thermoplastic composites is a versatile and efficient method of production. It allows for the creation of complex shapes and profiles with precise control over dimensions.

2.4. APPLICATIONS

2.4.1. TPCS Applications

Thermoplastic composites have a wide range of applications in various industries due to their enhanced mechanical properties and increased performance. These composites are particularly useful in industries such as aerospace, automotive, sporting equipment, and construction, where strong and lightweight materials are needed [24]. In the aerospace industry, thermoplastic composites are used for manufacturing aircraft components such as fuselage panels, wings, and interior parts.

i. Aerospace Industry

Thermoplastic composites have made significant advancements in the aerospace industry, primarily due to their superior reinforcing properties and ease of handling. These composites offer high specific stiffness and specific strength, making them ideal for use in aircraft manufacturing [25]. They are used in the production of structural components such as aircraft fuselage panels, wings, and interior parts. In addition to their lightweight and strong properties, thermoplastic composites also have excellent resistance to corrosion and fatigue, making them highly suitable for aerospace applications. Furthermore, the use of thermoplastic composites in aerospace helps achieve weight reduction, leading to reduced fuel consumption and transportation costs [26].

ii. Automotive Industry

The automotive industry is another sector where thermoplastic composites find extensive use. Thermoplastic composites offer several advantages in automotive applications.

They are lightweight, which helps improve fuel efficiency and reduce emissions. Additionally, thermoplastic composites have high strength-to-weight ratios and stiffness-to-weight ratios, making them suitable for structural components in vehicles [27]. Moreover, thermoplastic composites have excellent impact resistance and durability, which contribute to improved safety in automobiles. Furthermore, composites based on thermoplastic resins are preferred in the automotive industry due to their ease of processing, large-scale production capabilities, and the possibility of recycling [28].

iii. Renewable Energy

Thermoplastic composites also have applications in the renewable energy industry.

In the field of renewable energy, thermoplastic composites are used in the production of wind turbine blades. These composites offer the necessary strength and durability required to withstand the harsh environmental conditions and loads experienced by wind turbine blades. Furthermore, thermoplastic composites provide the advantage of easy maintenance and repair, which is crucial for the longevity and performance of wind turbines.

iv. Construction Material

Thermoplastic composites are also utilized in the construction industry for various applications. For instance, thermoplastic composites are used in the production of construction materials such as pipes, panels, and reinforcements. The use of thermoplastic composites in construction materials offers several benefits. These composites have high strength and stiffness, which contributes to the structural integrity of buildings. Furthermore, thermoplastic composites are resistant to corrosion and degradation, making them suitable for outdoor applications. In addition, thermoplastic composites in the construction industry provide ease of installation and customization, allowing for efficient and cost-effective construction processes. The versatility and convenience of thermoplastic composites make them a popular choice in diverse applications, including the automotive and aeronaut industries, construction materials, and renewable energy sectors. Overall, thermoplastic composites have gained popularity in a wide range of industries, such as automotive, aerospace, construction, and renewable energy [29].

2.4.2. Recycled TPCs applications

Recycled thermoplastic composites find a wide range of applications, particularly in the automotive industry, due to their recyclability and various other advantages [30]. These advantages include high-speed processability, excellent stiffness, and strength-to-weight ratios, and their ability to meet strengthening environmental regulations. In recent years, the automotive industry has increasingly turned to recycled thermoplastic composites for various applications. For example, recycled thermoplastic composites have been successfully used in the manufacturing of seatback frames and battery storage for hybrid vehicles. Furthermore, the use of recycled thermoplastic composites in auto parts offers several benefits [31].

One benefit is the reduction in energy consumption and carbon emissions during the manufacturing process, as recycled thermoplastic composites require lower processing temperatures compared to traditional thermosetting composites [32]. This not only lowers production costs but also contributes to more sustainable composite materials. The recyclability of thermoplastic composites allows for the reuse and regeneration of materials, reducing waste and contributing to a circular economy. In addition, the manufacturing flexibility of recycled thermoplastic composites enables the production of complex shapes and designs, allowing for lightweight and improved fuel efficiency in vehicles[30]. Another advantage of using recycled thermoplastic composites in automotive applications is their ability to enhance the crash performance and safety of vehicles. By absorbing and distributing impact energy more effectively than traditional materials, recycled thermoplastic composites can improve the overall structural integrity of automotive components. Figure 12 presents some typical applications of recycled TPCs.

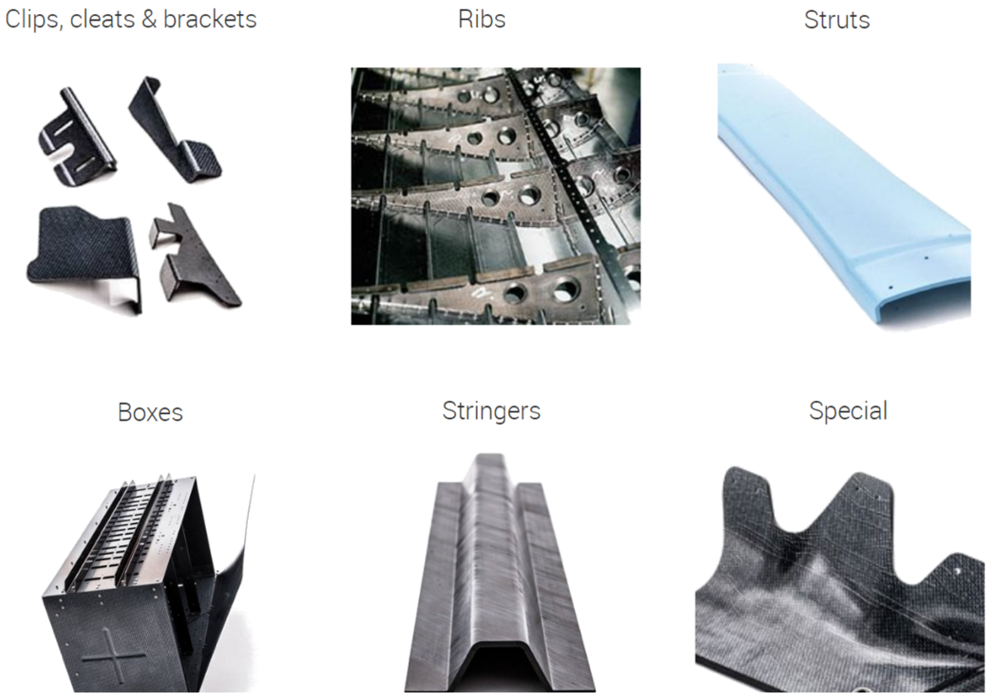

Figure 12 Examples of Applications of TPCs taken from [20]

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3. METHODOLOGY

The methodology considered for the execution of this work followed the four steps detailed in Figure 13. The first step consisted of the production of a composite plate (CFRP) via VARI-Vacuum Assisted Resin Infusion. The plate was cut into several parts and mechanically ground. The obtained recyclate was compounded with ABS in a single screw extruder and mixed manually. After the obtained composite material goes through hot compression, and lastly the plates processed go through physical and mechanical characterisation considering TGA, DSC, Capillary Rheometry and mechanical tests, although not considered in the present thesis, the material data obtained in the material test campaign, can be used for material constitutive modelling operations, towards the definition of the obtained materials in the context of injection moulding simulation.

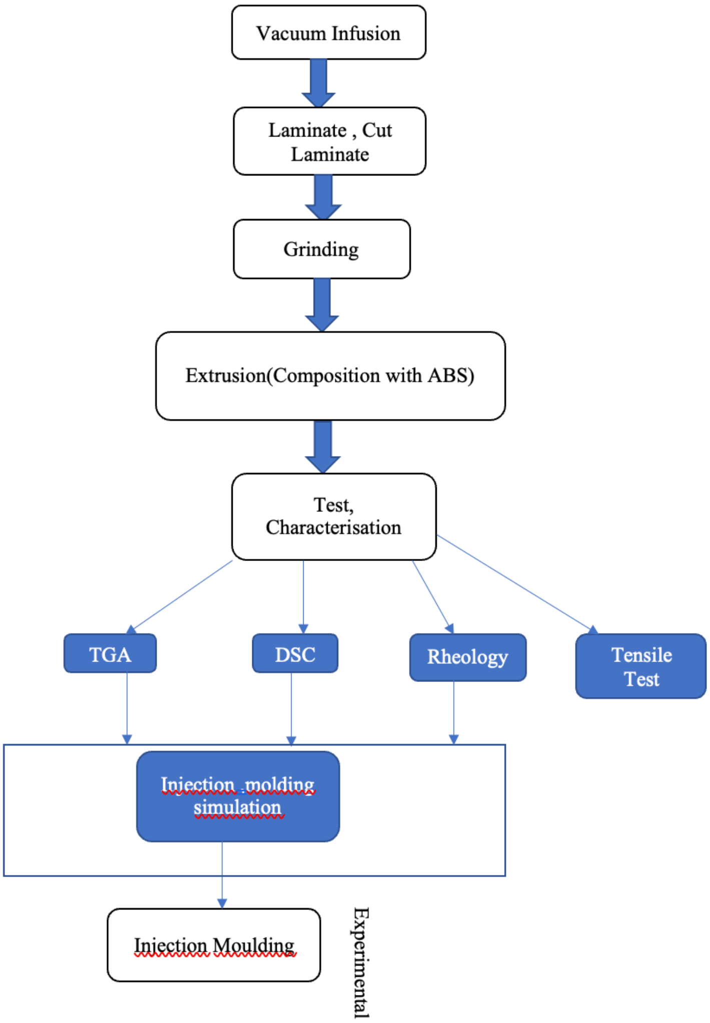

Figure 13 The overview of the methodology steps of this thesis

3.1. VACUUM ASSISTED RESIN INFUSION

The Vacuum Assisted Resin Infusion (VARI) is a technique that uses vacuum pressure to drive resin into a laminate. Dry materials are laid into the mould and the vacuum is applied before resin is introduced. The resin is then injected through one end of the part simultaneously with the application of a vacuum at the other. During the process, the vacuum has the function of directing the resin front, in addition to eliminating possible porosities caused by air and volatiles released during the curing reaction of the part. Once a complete vacuum is achieved, the resin is sucked into the laminate via carefully placed tubing [16]. This process is aided by an assortment of supplies and materials as it has been depicted in Figure 14.

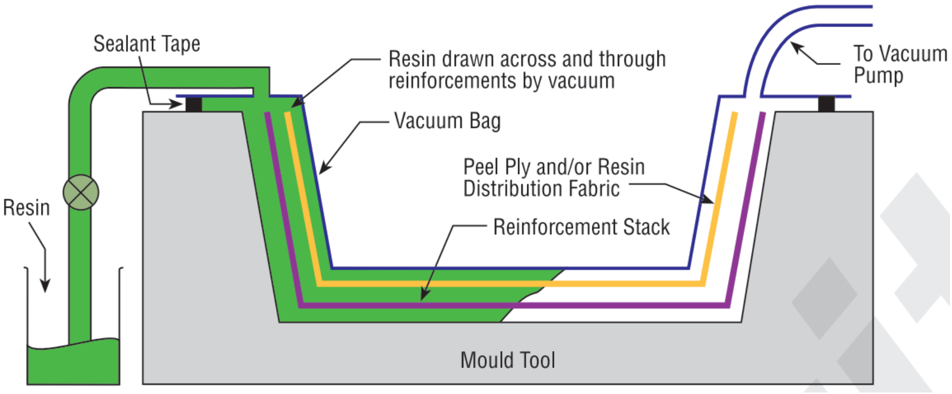

Figure 14 VARI-Vacuum Assisted Resin Infusion process taken from [33]

3.1.1. Manufacturing Process

In this study, the VARI technique was used to infuse the thermoplastic resin (ELIUM 188XO) into a fibrous carbon reinforcement of 10 plain layers of carbon fibres with orientation 0/90º. During the VARI process, ELIUM resin was used as the matrix material for impregnating the fibre preform. Its low viscosity and good wetting properties allow for easy and uniform resin flow, resulting in a high-quality composite. Once the layup process was done, the preform was covered with a vacuum bagging film, ensuring airtight sealing around the edges and a vacuum pump was connected to create a vacuum inside the bag, which helps remove air and facilitates resin infusion. ELIUM resin was then introduced into the layup using the VARI technique. This involved placing a resin feed line connected to a bucket containing ELIUM resin on top of the layup. The vacuum pressure was then applied to draw the resin through the preform, impregnating it evenly. After the resin infusion, the part was left to cure at room temperature since the thermoplastic resin ELIUM cures at room temperature (22 °C).

Once the part had cured and solidified, it went through the post-processing step of demoulding, allowing the obtaining of a composite laminate composed of 10 layers, with a final thickness of 2 mm.

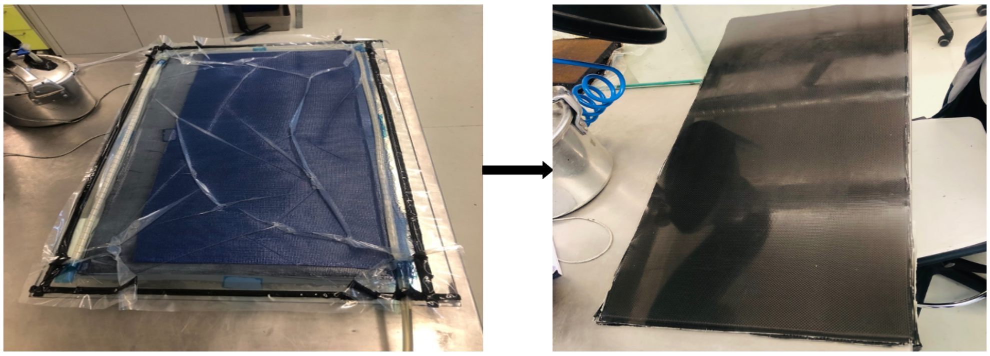

Figure 15 post-infusion process and de-moulding

The kit used for this process consists of a vacuum pump, pressure pot, inlet, and outlet hoses for infusion of resin, peel plies, breather, mesh flow, and spiral tube, to name but a few. Figure 15 shows the infusion of the composite part. Table 2 summarizes the materials and auxiliary equipment used to produce the composite panel.

Table 2 Materials used for VARI.

Material	**Quantity**
Layers of Carbon fibre (900mm x 400mm)	10
Thermoplastic resin (ELIUM 188XO)	862 g
Peroxide Benzyl	25,9 g
Infusion line clamps	2
Vacuum Bag	1
Vacuum pump	1
Vacuum lines	1

3.2. MECHANICAL RECYCLING

The composite panel was cut into several parts and mechanically ground to obtain a composite granulation (recyclate). The grinding of carbon fibre-reinforced polymer (CFRP) to make flakes, reduces the composite material into smaller flakes, reducing the fibre length. To achieve this, surfacing grinding methods were used. Surface grinding is a common method where the CFRP is placed on a grinding wheel or surface, and the material is removed gradually until flakes of the desired size are obtained. It is important to consider the process and parameters for achieving the desired results. Samples and grinding machine steps used are shown in Figure 16.

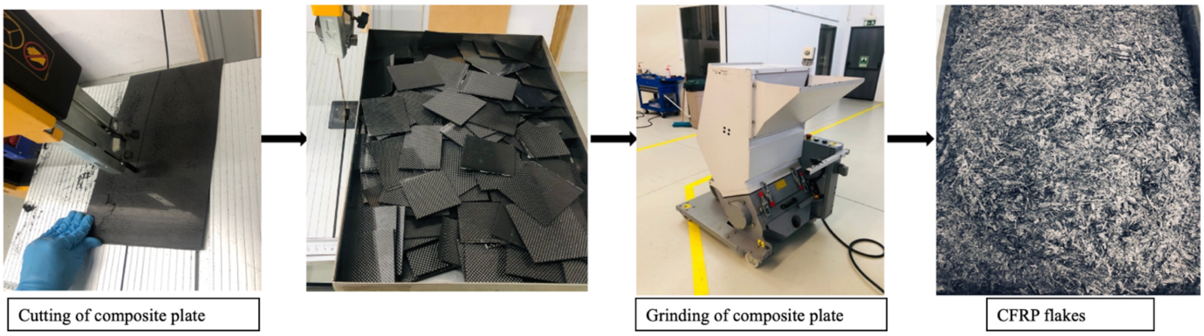

Figure 16 Cutting laminates and grinding.

3.3. COMPOSITION OF RECYCLATE WITH ABS

The recyclate obtained was combined with ABS thermoplastic material. The use of CFRP combined with ABS offers several advantages. Firstly, the addition of carbon fibres enhances the mechanical properties of the composite material. The carbon fibres provide increased stiffness and strength, enabling the production of functional parts that can withstand higher loads. Furthermore, the incorporation of thermoplastic matrixes allows for greater design flexibility within the composite recyclate context. This flexibility is achieved through the ability to vary the composition, fibre volume and distribution of carbon fibres within the composite matrix, allowing for tailored mechanical properties based on specific application requirements.

3.3.1. Mixing

ABS (Acrylonitrile Butadiene Styrene) and CFRP composite are two different materials, with ABS being a thermoplastic polymer and CFRP being a reinforcing material with reinforcement fibres embedded in a PMMA matrix. Although specific compatibility data between ABS and

PMMA was found, it was discovered that ABS and PMMA (ELIUM) were compatible. To ensure even distribution and optimal reinforcement in the study, 50% virgin ABS was mixed with 50% CFRP material after the grinding process had occurred, leading to a 1:1 ABS/CFRP material ratio. To investigate the difference in mechanical properties, the same method was conducted by doubling the amount of ABS in the CFRP composite reaching a 2:1 ABS/CFRP ratio. As shown in Figure 17 1kg of the CFRP composite was mixed with 1kg of virgin ABS leading to a 1:1 ABS/CFRP ratio composite material.

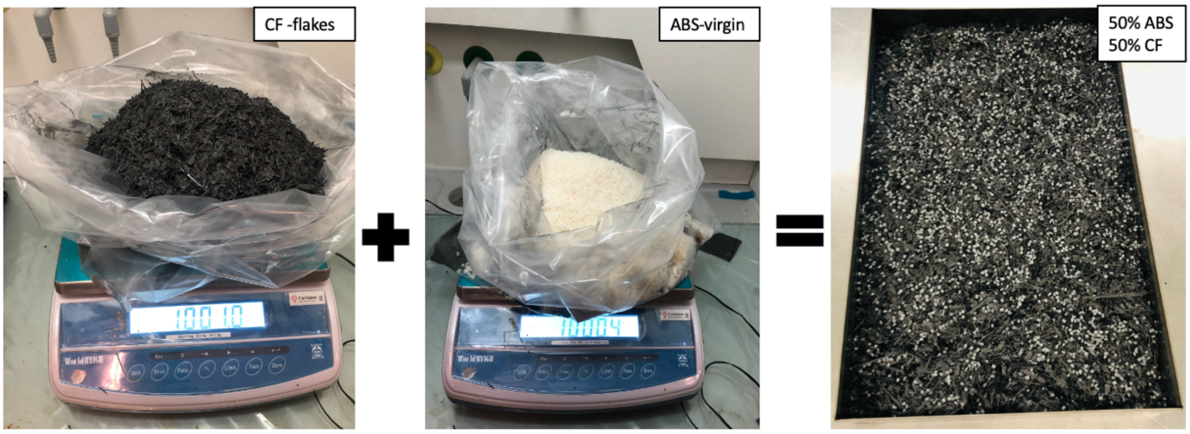

Figure 17 Mixing of ABS and CF flakes.

3.3.2. Material Composition

The mixing process was carefully done to guarantee a good mixture of both CFRP and ABS matrixes. Once the mixture was ready, it was fed into an extruder where it underwent melting and homogenization. The extruder, equipped with a barrel and heating zones, used a screw to convey the material forward while applying heat and pressure to melt it. Finally, the molten mixture was forced through a die with a specific cross-sectional shape to form a continuous profile. It should also be mentioned that a portion of the mixture ABS/CFRP obtained was not homogenised in the extruder and, alternatively, homogenized during hot compression for the evaluation of the influence of the homogenization technique on the mechanical properties.

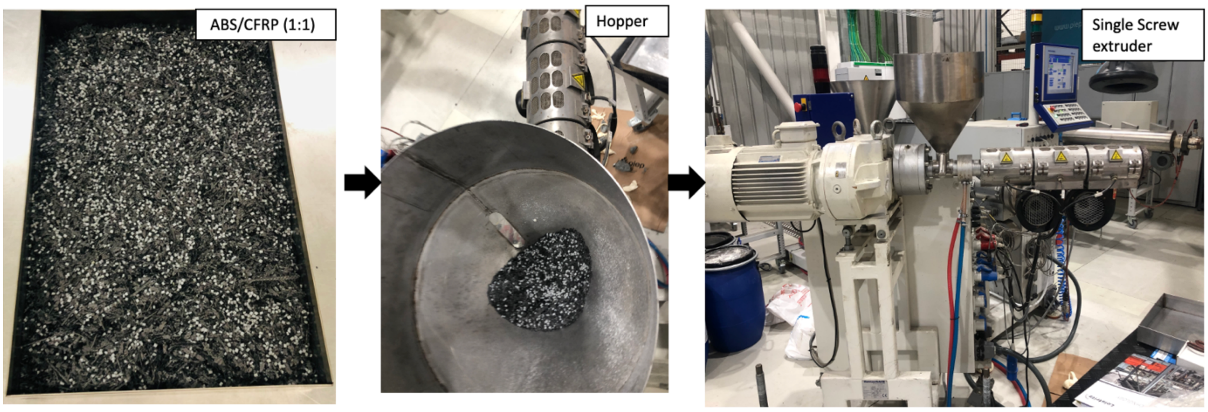

Figure 18 Extrusion Process

3.3.3. Hot Compression Moulding

After the material composition process is completed, the materials go through compression moulding, the layup assembly is placed into a compression mould cavity. The compression force applied to the mould helps to consolidate the ABS/CFRP material, ensuring good interfacial bonding and uniform distribution of the fibres. Compression moulding was performed in a hot press using a mould preheated.

The material was first placed in a hot open mould for about 10 minutes at the temperature of 180 °C with intermediate pressure and then the mould was closed, and pressure was slowly applied until it reached a final value of 50 bars. After reaching the final pressure, the plate was allowed to consolidate for 5 minutes before cooling to ambient temperature. The cooling process allows the ABS/CFRP matrix to solidify and bond with the carbon fibres, resulting in a strong and durable composite structure. Once the cooling stage was complete, the mould was opened, and the composite part was removed. The part is then allowed to cool down to room temperature before further processing or testing. For the hot compression moulding process, a hot plate press with a dimension of 200x200mm was used as shown in Figure 19.

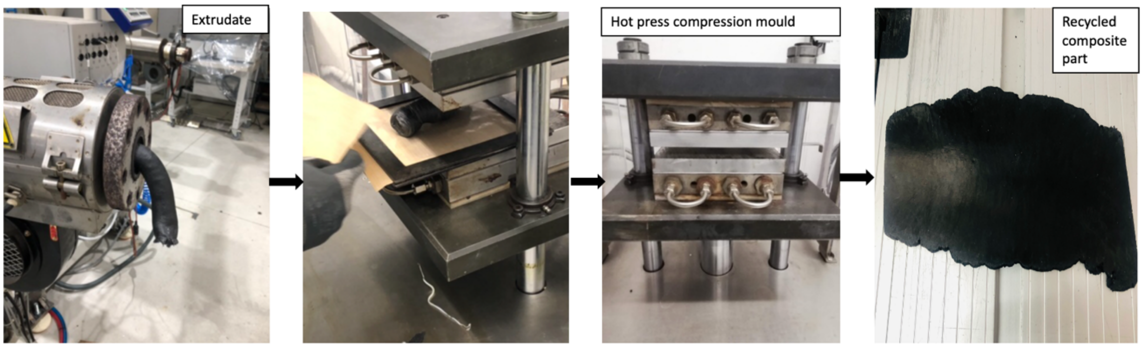

Figure 19 Direct compression moulding process.

3.4. TEST CHARACTERIZATION

3.4.1. Mechanical Test

Tensile tests were carried out at room temperature, and the experimental procedure and the sample dimensions were defined according to ISO14125 The mechanical response of the recycled composite material was evaluated through flexural tests, following the standard ASTM D7264. The tests were carried out using a universal testing machine (UTM). The specimen was placed in the machine's grips and subjected to increasing force until it reached failure. During the test, the machine measures the force applied and the displacement of the specimen. Both Young Modulus and Stress at Break were measured for 10 samples, 5 samples with the composition of 1:1 ABS/CFRP composite and 5 samples with the 2:1 ABS/CFRP ratio composite, at room temperature (22ºC). The experimental setup considered is seen in Figure 20 which shows the composite part that went through the extrusion process and then was compressed in a hot compression moulding. Figure 21 shows a composite part that was mixed through direct compression moulding as can be seen the composite part has less homogenisation.

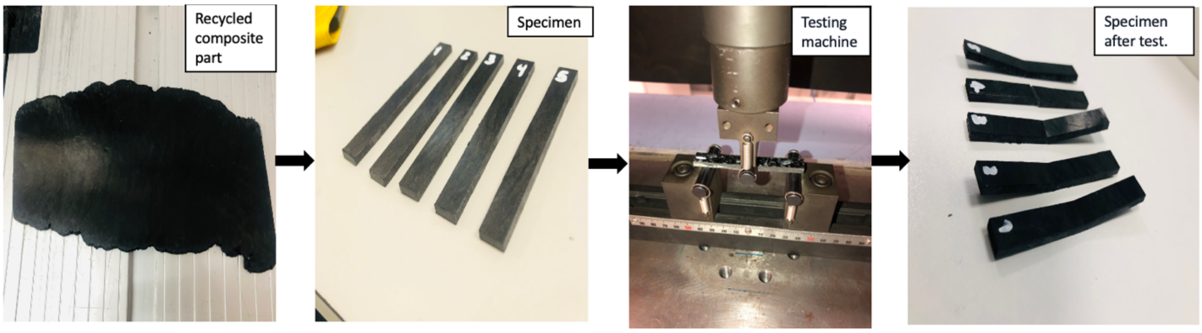

Figure 20 Experimental Setup for Mechanical Characterization ABS/CFRP (processed through extrusion and went to extrusion).

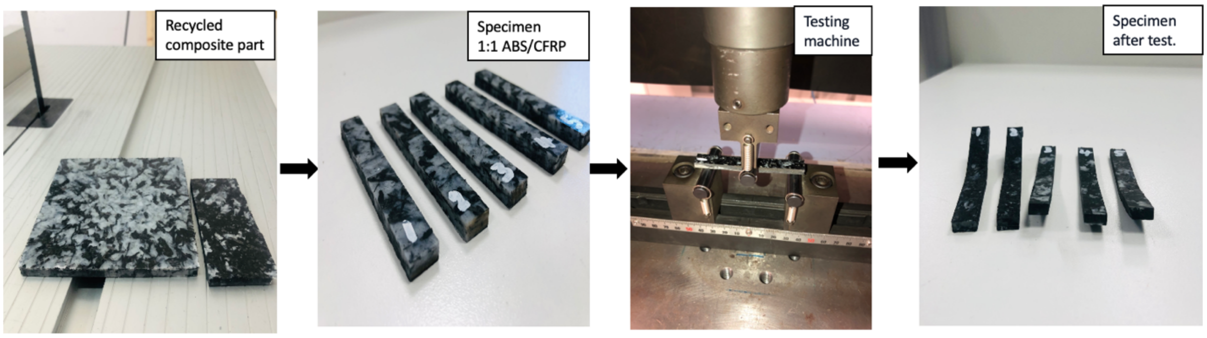

Figure 21 Experimental Setup for mechanical characterization ABS/CFRP (processed through direct compression moulding).

The following equation gives the flexural stress of:

Equation 1 Equation To calculate the stress of the specimen.

$$\sigma_f = \frac{3FL}{2bh^2}$$

σ_f is the flexural stress, in megapascals (MPa).

F is the load in newtons (N).

L is the span, in millimetres (mm). (64 mm)

h is the thickness of the specimen, in millimetres (mm).

b is the width of the specimen, in millimetres (mm).

To calculate the strain on the outer surface of the specimen Equation 2 was used.

Equation 2 To calculate the strain of the specimen.

$$\varepsilon = \frac{6sh}{L^2}$$

ε is the strain, in (%)

h is the thickness of the specimen, in millimetres (mm).

L is the span, in millimetres (mm). (64mm)

s is the displacement, in (mm)

3.4.2. Fibre length

To determine the length of the fibres, the carbon fibres were extracted ABS/CFRP matrix. A burning treatment was conducted in an inert atmosphere at 700 °C to remove the polymer matrix. The resulting ash was used to extract single fibres, which were then dispersed in water and dried. A sample containing fibres was prepared and placed on a microscope slide, which was inserted into an optical microscope equipped with a digital imaging system (see Figure 22). The stereoscope was adjusted to a suitable magnification level to ensure that individual fibres were distinguishable. Images of the fibre distribution and length were captured using the microscope and processed using image analysis software, which allowed for automated measurement of fibre length. To calibrate the system, the scale of the image was determined by capturing an image of a calibration slide, with known dimensions, or by using a stage micrometre. Once the scale was established, the software could accurately measure distances within the image. This method was applied to two different compositions: 1:1 ABS/CFRP and 2:1 ABS/CFRP composite. Additionally, morphological analyses were conducted on sample sections taken near the fracture surfaces using the optical microscope.

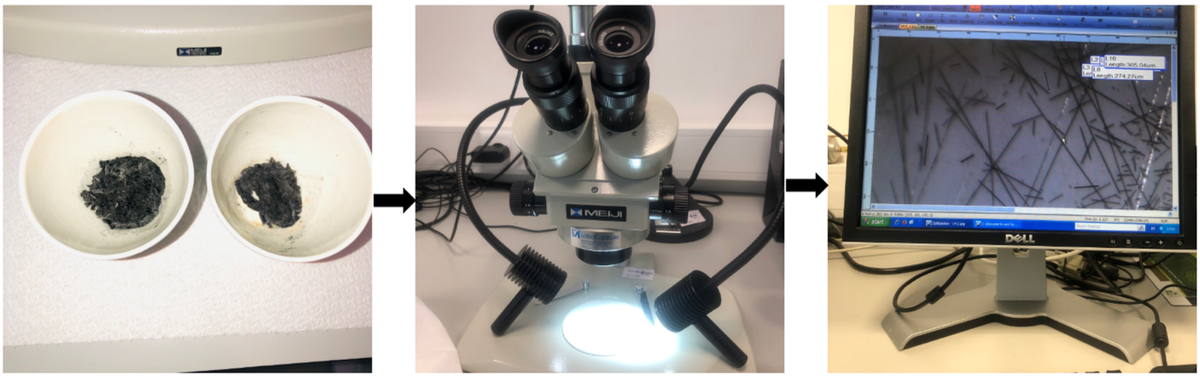

Figure 22: Method of calculating the fibre length through optical stereoscope.

3.4.3. DSC

The experiment involved analysing the properties of test specimens through Differential Scanning Calorimetry (DSC) tests as shown in Figure 23. The DSC tests were conducted on ABS, 1:1 ABS/CFRP and 2:1 ABS/CFRP compounds as-received pellets and extruded materials, to determine their glass transition (T_g) and enthalpies. The sample masses ranged from 2 to 3 mg, and the tests were performed using a DSC following ISO-11357-3. To prevent contamination of the test environment during heating and cooling sequences, aluminium T_{zero} containers were used to seal the test specimens. The temperature was first equilibrated at 30 °C and held isothermally for 5 minutes. Then, the temperature was gradually increased from 30°C to 300 °C at a rate of 10 °C/min, and the DSC instrument measured and recorded the heat flow as a function of temperature during the heating cycle. The data gathered helped generate a DSC thermogram, which represented the changes in the material's heat capacity as it underwent various thermal transitions such as glass transition and, eventually, material degradation.

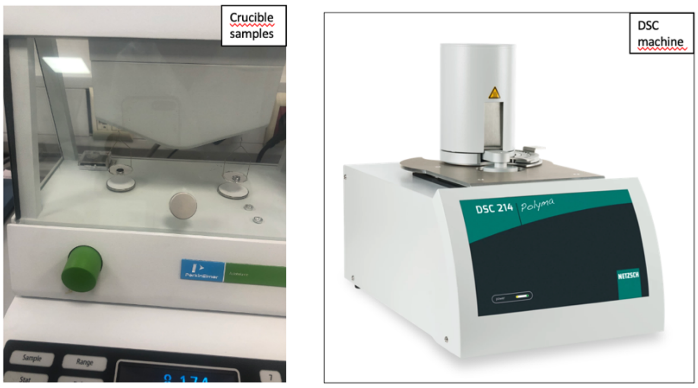

Figure 23 DSC experiment setup

3.4.4. TGA

Thermogravimetric analysis (TGA) and differential thermogravimetric analysis (DTGA) were performed on a TGA 55 instrument as shown in Figure 24, to determine the temperature at which degradation begins and the weight percentage of the filler/fibre in the matrix materials. The TGA instrument was calibrated for temperature, heat flow, and weight according to the manufacturer's guidelines. The TGA tests were conducted by heating the samples from room temperature (30°C) to 800°C at a heating rate of 10°C/min as per the ASTM Standard E1131-20. A nitrogen gas flow rate of 60 ml/min was used for the sample purge, while the balance purge was set at 40 ml/min, delivered at 20 psi. The samples, weighing between 3 to 5 mg, were placed in a high-temperature platinum pan, and subjected to degradation as a function of temperature, with their mass loss recorded by the TGA 55's TRIOS software. The temperature at which 1% weight loss occurred was considered as the degradation onset temperature (DOT). The weight percentage loss was plotted concerning temperature using the experimental data.

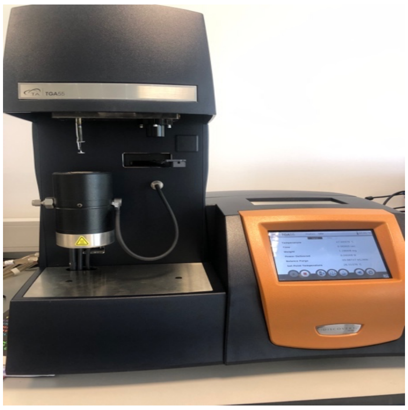

Figure 24 TGA 55 instrument

3.4.5. Capillary Rheology

The granules obtained from the composition process were dried and freed from any moisture or contaminants that could affect the rheological measurements. The instrument was set up according to the rheometer manufacturer's instructions. This involved attaching the appropriate die and selecting the desired measurement parameters, such as temperature at 180 °C, pressure, shear rate, and shear stress range. Calibrate the instrument using a known reference material with well-characterized rheological properties. This calibration helps ensure accurate measurements of the ABS and CFRP granules. The temperature of 180 °C was set for the test. This can be done using built-in heating or cooling mechanisms within the capillary rheometer. First, the granules were loaded into the capillary rheometer's sample chamber. It was ensured that the sample was uniformly distributed and removed any air bubbles or voids that may be present. During the test, the capillary rheometer measures the pressure drop or torque (shear stress) as a function of the shear rate or shear stress. This data is then utilized to generate flow curves and rheograms. These graphical depictions provide a visualization of the material's flow behaviour and viscoelastic properties.

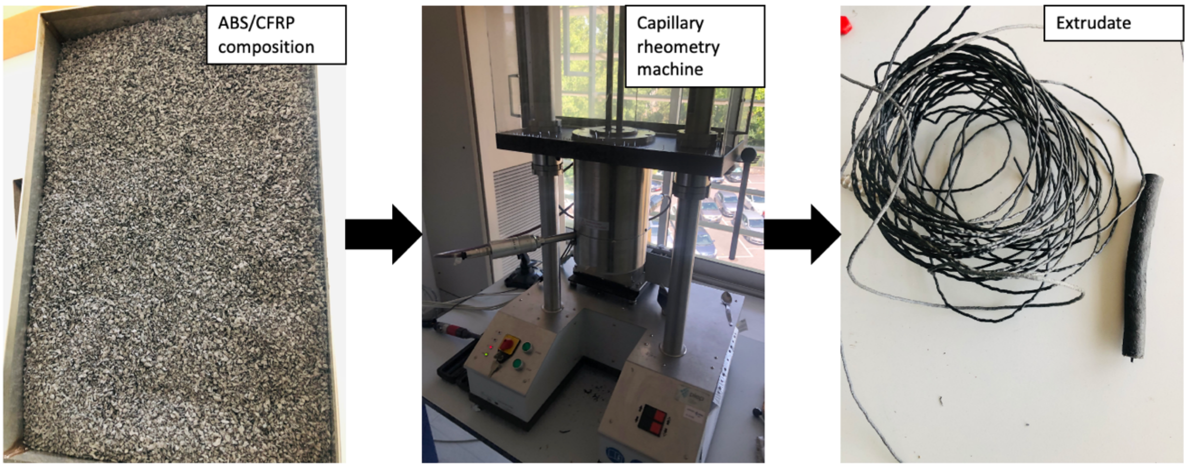

Figure 25 Rheometry Setup

4. RESULTS AND DISCUSSION

The study aimed to assess how mechanical recycling affects the ultimate properties of thermoplastic composites utilizing an Elium (188XO) polymeric matrix reinforced with short carbon fibres. The resulting composite materials could be utilized for various automotive components. By comparing the mechanical properties of Thermoplastic Polymer Matrix composites (TPMC) to those of traditional composite materials, including strength, stiffness, and impact resistance, we could gain insights into the performance characteristics and potential benefits of TPMC in different applications.

4.1. FLEXURAL TEST

Flexural tests for the thermo-compressed specimen were conducted and evaluated based on the D7264 ASTM standard. This ASTM standard was used to determine the flexural stiffness and strength properties of the developed polymer matrix composites.

In our experiment, we prepared specimens of the ABS/CFRP composite according to standard test procedures. The specimens were then subjected to a controlled force until they reached failure. The applied force and corresponding elongation were recorded to quantify the mechanical characteristics of the composite. Flexural modulus and Stress at Break of the composites were collected and statistically analysed. Mean values and standard deviation are indicated in Table 3 and Table 4. The results obtained from the flexural test of the 5 specimens considered, their maximum stress as well and the average and standard deviation of the results are summarized in Table 3 and Table 4.

Table 3 Young modulus and Stress 1:1 ABS/CFRP

Specimen	Young Modulus (MPa)	Maximum Stress (MPa)
1	7382.86	134.260
2	10881.4	139.940
3	14222.1	169.967
4	6832.20	145.265
5	12245.7	139.598
Average	**10312.9**	**145.806**
Standard deviation	**3163.90**	**14.0562**

Table 4 Young modulus and Stress 2:1 ABS/CFRP.

Specimen	Young Modulus (MPa)	Maximum Stress (MPa)
1	3627.84	74.3344
2	3281.13	68.6311
3	3002.31	70.9745
4	3455.61	75.0081
5	4080.43	75.8390
Average	**3489.46**	**72.9574**
Standard deviation	**403.193**	**3.04359**

To calculate the modulus of elasticity (Young Modulus) Equation 3 was used, From ISO14125 it is stated that the measurement of the flexural modulus can be determined by calculating the deflections which correspond to the given values of flexural strain εf' = 0,0005 and εf'' = 0,0025, by the following equation:

Equation 3 Young Modulus

$$E = \frac{\sigma_{0,25\%} - \sigma_{0,05\%}}{0,0025 - 0,0005}$$

$\sigma_{0,25\%}$= 18,96 M Pa

$\sigma_{0,05\%}$=4,07MPa

$E =$ 7442,91 MPa = 7,4 GPa

The Young modulus was found to be 7442,91 MPa.

Table 5 are summarized the values obtained for the Young Modulus, considering the different material composition and homogenization techniques (extrusion and hot compression) and composition ratios.

Table 5 Young modulus of the specimen based on the homogenization method.

Specimen	Young Modulus (GPa)
Extrusion ABS/CFRP 1:1	7,442
Compression ABS/CFRP 1:1	8,478
Compression ABS/CFRP 2:1	3,368
Extrusion ABS/CFRP 2:1	3,574
Average	**5,716**
Standard deviation	**2,725**

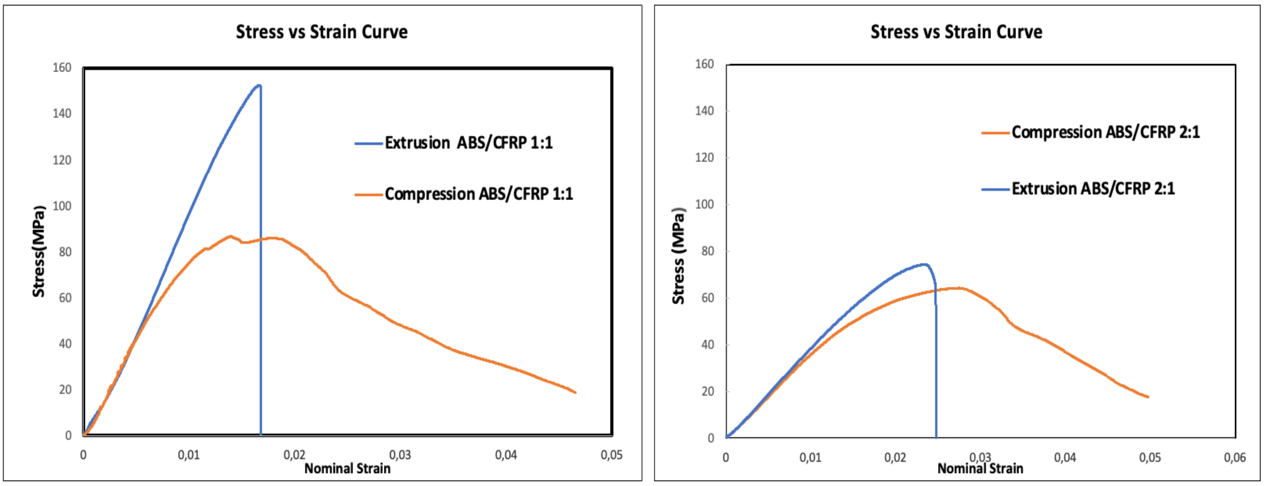

Figure 26 Stress and strain curve with ABS and CFRP (1:1 and 2:1)

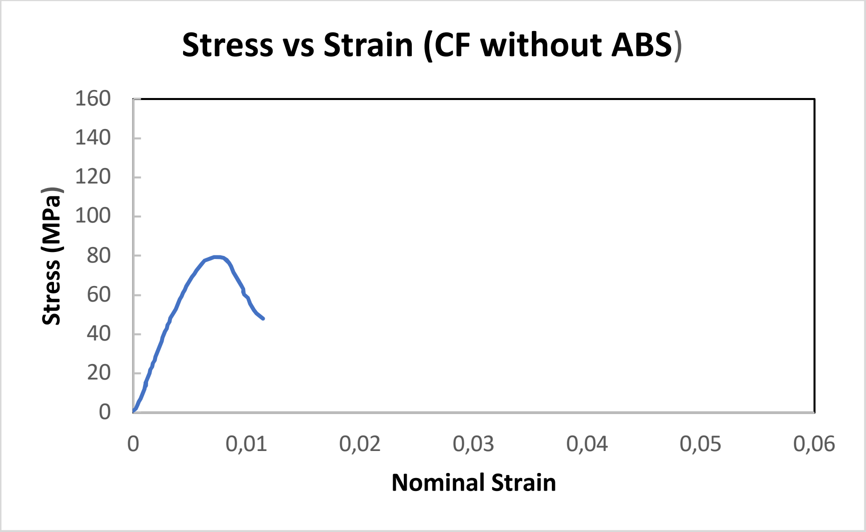

Figure 27 Stress and Strain curve for the CFRP recyclate (0% ABS)

The curve Figure 26 shows, for the raw ABS/CFRP material obtained from the homogenization process with extrusion, a nearly linear behaviour until failure and clearly shows that the specimens fail immediately at a maximum, revealing a fragile rupture. The material behaviour obtained with raw material not homogenised via extrusion and hot compressed directly showed a more ductile behaviour. This behaviour could be explained by the less homogeneity obtained in the samples produced, which can contribute to the appearance of voids and, consequently, stress concentration spots leading to a precocious and more progressive failure of the ABS/CFRP composite material. Figure 27 depicts the mechanical behaviour of the CFRP material without the incorporation of ABS. The Young Modulus obtained is higher when compared to the elasticity modulus obtained for the 1:1 and 2:1 ABS/CFRP formulations, due to the higher fibre volume of this material composition, which has only PMMA in its composition.

The presence of carbon fibre reinforcement in the thermoplastic matrix had a significant impact on the mechanical properties of the composite. The addition of carbon fibres (an increase in volume fibre) enhanced the overall strength and stiffness of the material, leading to an increase in the Stress at Break and Young's modulus. This is visible for the 1:1 ABS/CFRP material which, due to the increase of fibre volume, reaches maximum values of Young Modulus and Stress at break. It should be noticed that the Young Modulus for the 1:1 ABS/CFRP composite is almost double the Young Modulus obtained for the 2:1 ABS/CFRP composite. For the same formulation, the maximum strain values were higher in the composites manufactured by compression a plastic deformation, not observable in the extruded composites. This is because when a material is compressed, it is subjected to forces that result in a more uniform distribution of molecules or particles within the material. This compression can align the particles and create a more homogeneous structure, leading to isotropy. As a result, compressed materials tend to have consistent physical properties, such as density, strength, and conductivity, in all directions.

In contrast, extruded materials are formed by forcing the material through a die or nozzle, often resulting in a directional alignment of the particles or molecules. This alignment can create anisotropic properties, where the material exhibits different characteristics depending on the direction of measurement. Hence fibre alignment in extrusion increases its strength and reduces its strain.

Young's modulus describes the stiffness or rigidity of a material. The ABS/CFRP composite displayed a higher Young's modulus than pure ABS, indicating increased stiffness due to the presence of carbon fibre reinforcement. The carbon fibres act as a strengthening agent, preventing excessive deformation and improving the overall structural integrity of the material. Based on the models there was an increase of about 5 GPa, for the 1:1 ABS/CFRP material when compared with baseline ABS.

The flexural test also provided insights into the strain at break or elongation at break of the ABS/CFRP composite. The addition of carbon fibres (increase in volume fibre) resulted in a slight reduction in the elongation at break compared to pure ABS. This decrease in elongation suggests that the composite has a lower capacity for deformation before fracture when compared to a non-reinforced material.

4.2. FIBRE LENGTH (OPTICAL MICROSCOPY)

The optical microscopy fibre length test experiment of ABS/CFRP polymer was conducted to evaluate the dispersion and distribution of carbon fibres within the ABS/CFRP matrix, as well as to determine the average fibre length. It should be stated that, for this study. only the composite materials homogenised through extrusion were considered. Using the optical microscopy images, the length of individual fibres was measured. Approximately, 500 fibres were measured to obtain a representative average fibre length value. The measured fibre lengths were used to calculate the average fibre length, which was equal to 370 µm for the 1:1 ABS/CFRP composite and equal to 377 µm for the 2:1 ABS/CFRP material.

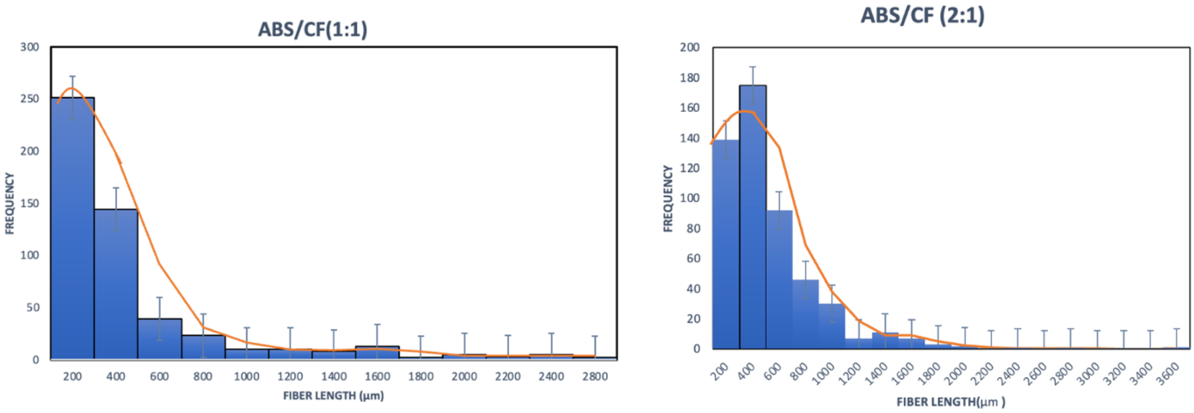

Figure 28 Fibre length distribution (histogram)

The optical microscopy images in Figure 29 reveal the dispersion and distribution of carbon fibres within the ABS matrix. A dispersed and evenly distributed network of fibres throughout the polymer matrix can be seen for 1:1 ABS/CFRP composition compared to the one that had less fibre volume (2:1 ABS/CFRP).

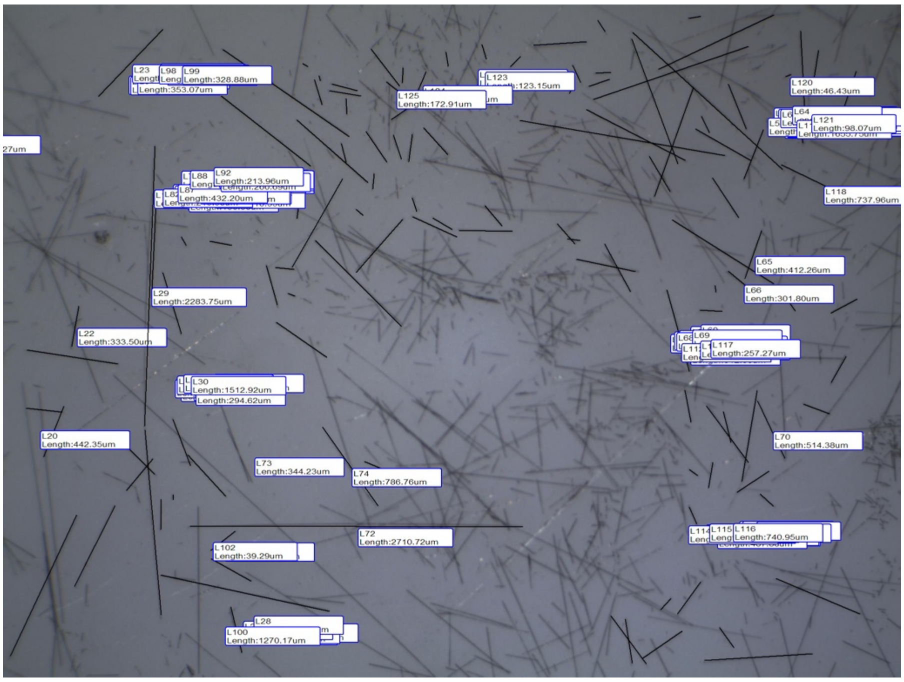

Figure 29 Optical image of calculating fibre length.

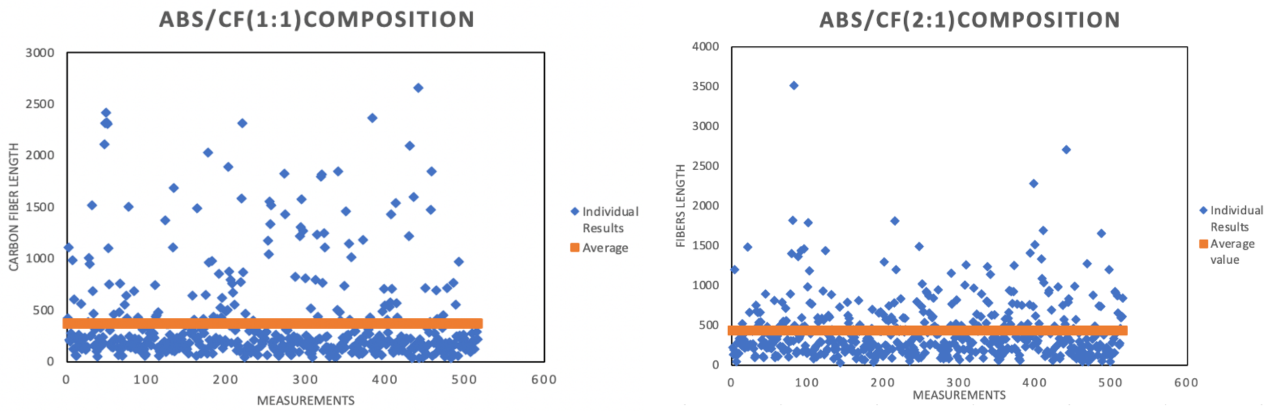

Figure 30 Fibres length distribution

It was noticed that as the ABS incorporation ratio increases the fibre length increases. It was expected that an increase in the amount of polymeric matrix would result in a less severe thermomechanical loading scenario, inside the extrusion cylinder (leading to the minimization of shear stress), leading to a less aggressive fibber cutting phenomena and, consequently, to the maximization of fibre length. The presence of polymer matrices facilitates better interfacial interaction between the fibres and the matrix, improving the transfer of stress and load-bearing capabilities. It can also help prevent fibre aggregation and promote more uniform distribution.

4.3. DSC

The DSC thermogram of the ABS/CFRP composite exhibited several characteristic peaks and transitions as seen in Figure 31 and Figure 32. The following observations were made:

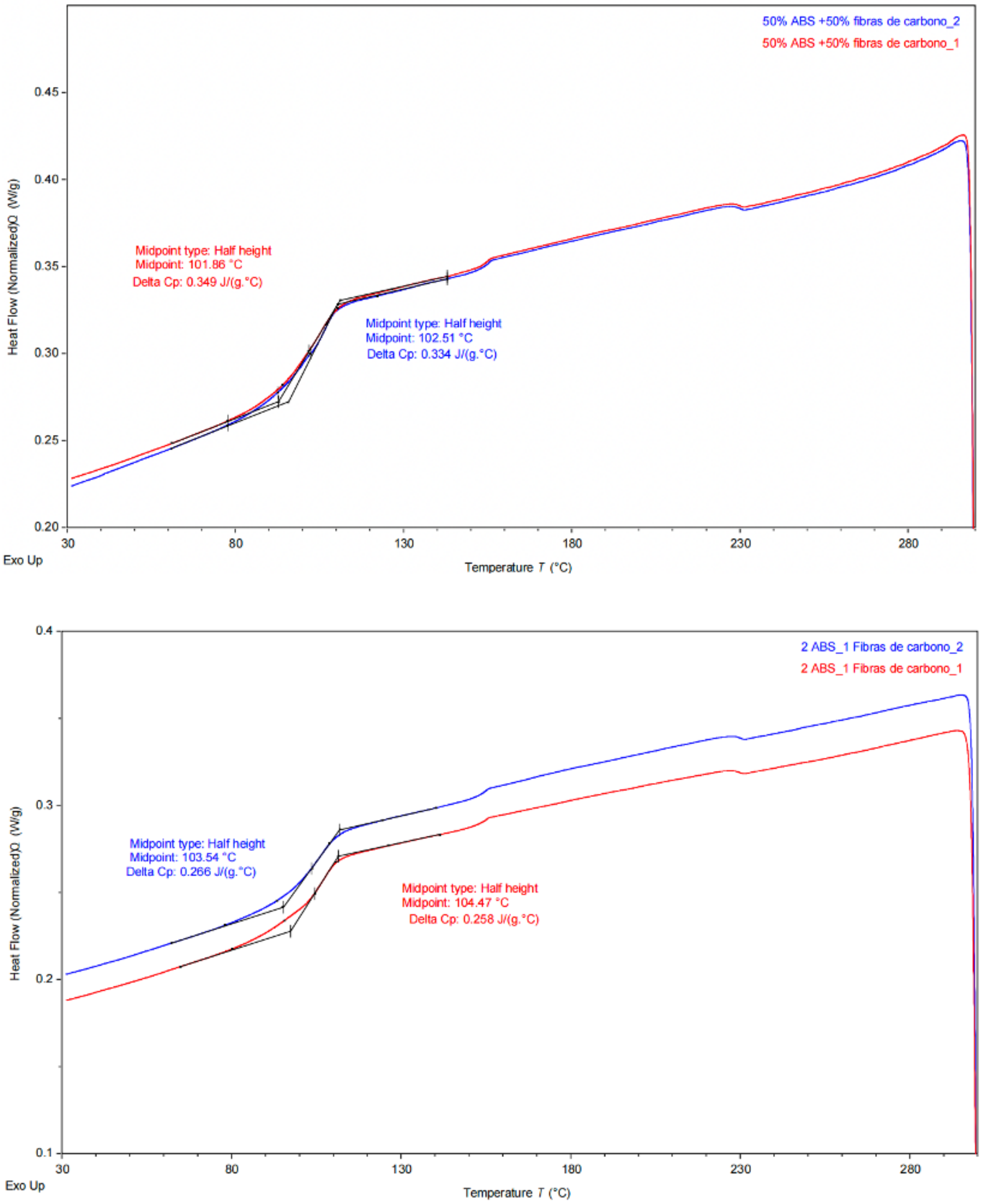

Figure 31 DSC curve of crystallisation

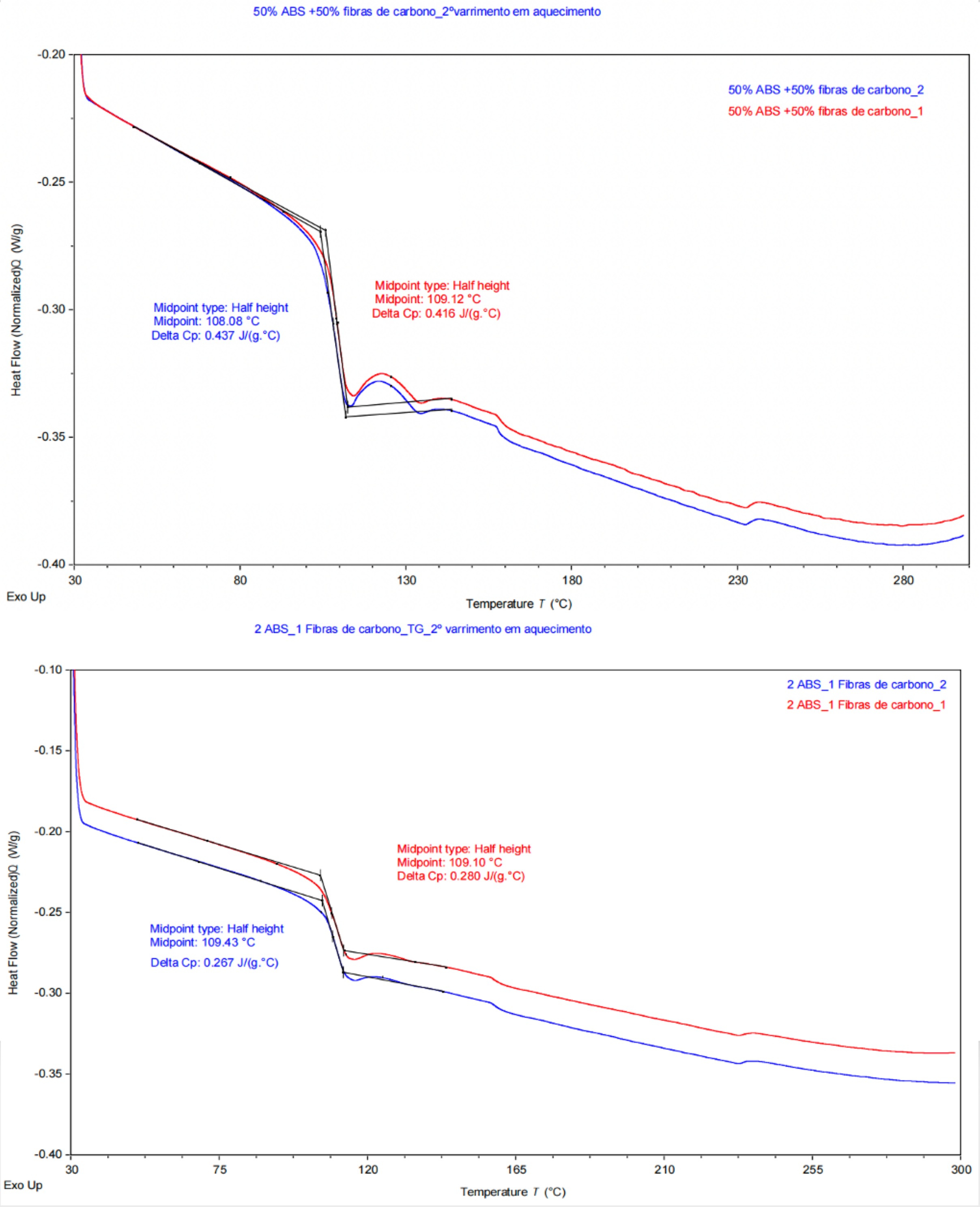

Figure 32 DSC curve for the heating process

1) The DSC curve displayed a distinct glass transition peak (T_g) for the ABS matrix. The T_g of ABS is typically around 105°C. However, the presence of carbon fibres and PMMA in the composite influenced the T_g, potentially shifting it slightly. The incorporation of carbon fibres in the ABS matrix can influence the polymer chain mobility, which might also affect the T_g. The interaction between the carbon reinforcement and the ABS/CFRP matrix can restrict

molecular motion, potentially increasing T_g. Additional DMA tests would be beneficial to investigate this effect in more detail.

2) The DSC experiment can provide insights into the thermal stability of the ABS/CFRP composite. The onset of thermal degradation, reflected by an endothermic peak, can be determined. The presence of CFRP reinforcement might affect the thermal stability of the composite, potentially improving it due to the thermal properties of carbon fibres.

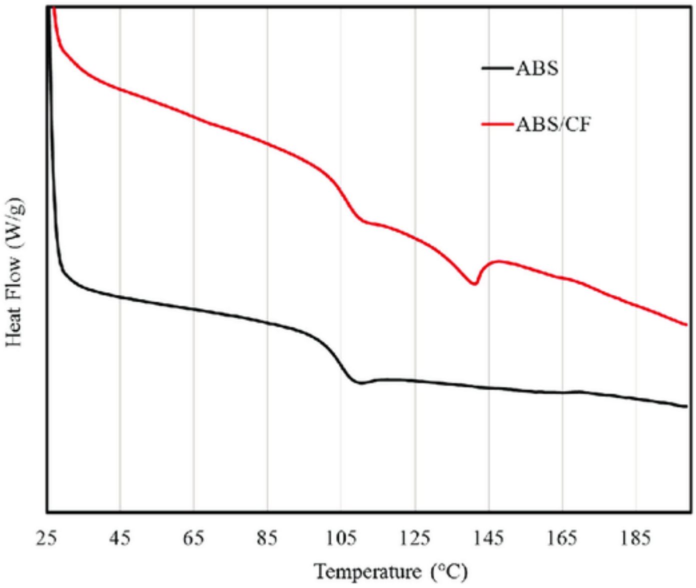

Figure 33 DSC thermograms of neat ABS and ABS-based composite taken from [34] *.*

Figure 33 shows the thermograms of DSC tests for ABS and ABS-based short 20 wt. % CF reinforced composite. In the case of neat ABS, there was a transition in the thermogram at 110 °C which is known as the glass transition temperature. It represents the transition from the glassy phase to the rubbery phase. As the ABS is amorphous, there is no definite melting temperature. The addition of CF to the ABS matrix had a slight impact on the polymer chain mobility. The Tg of 20 wt. % CF-based ABS was found at 105 °C which was lower than the neat ABS at 110°C. Increased chain mobility due to the conductive fibres within the matrix material reduces the glass temperature. In other words, the chopped fibres contributed to the enhancement of the thermal mobility of the polymer chain; thus, at a relatively lower temperature matrix, materials transformed from the glassy to the rubbery phase[34].

4.4. TGA

The thermogravimetric analysis (TGA) experiment of ABS with carbon fibre reinforcement polymer aimed to investigate the thermal stability and decomposition behaviour of the composite material. Through TGA, we can analyse the weight loss of the sample as a function of temperature. The TGA analysis was conducted using a heating rate of 10°C/min in an inert atmosphere.

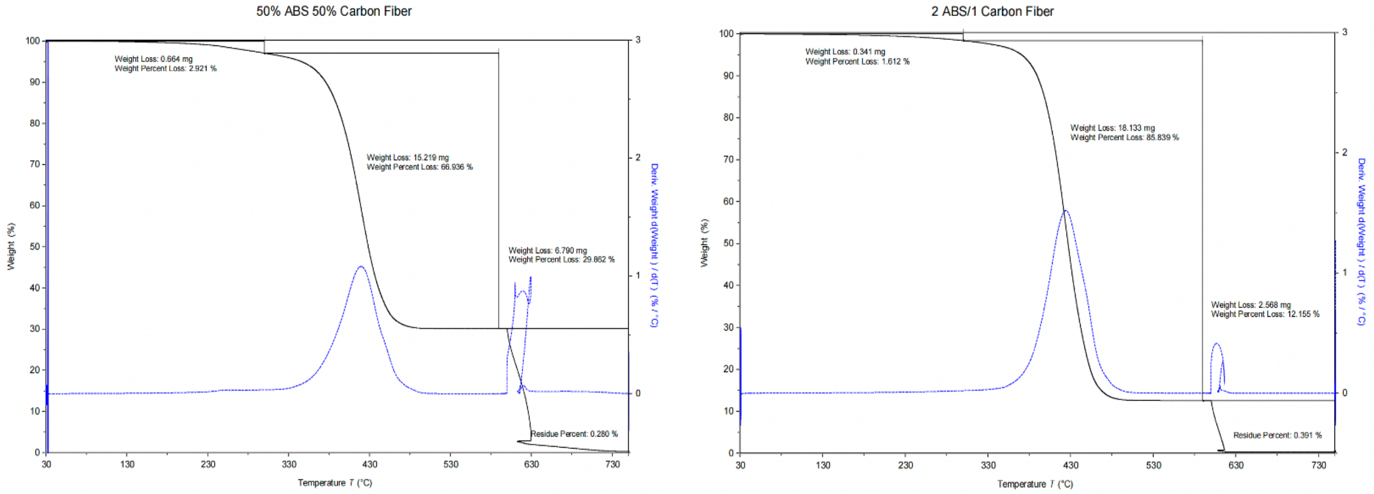

Figure 34 TGA thermal degradation curve

Figure 34 shows the TGA curve of the ABS/CFRP composite displayed two distinct weight loss regions. The first weight loss occurred at a relatively low temperature, corresponding to the evaporation of moisture and volatile substances absorbed by the sample. This initial weight loss accounted for less than 5% of the total weight loss. The second weight loss region, which took place at higher temperatures, was associated with the thermal degradation of the ABS/CFRP polymer matrix. As the temperature increased further, both organic components started to decompose, resulting in mass loss. The decomposition process involved various degradation reactions, such as depolymerization, chain scission, and the release of volatile organic compounds.

The residual weight is high on the composition of 1:1 ABS/CFRP weights 29% compared to 12% of 2:1 ABS/CFRP material. This is the prove that we have more fibre volume, in the first composition (1:1 ABS/CFRP), which can be correlated with the increase of young modulus and Stress at Break as shown in Section 4.1, for the 1:1 ABS/CFRP composite.

The TGA curve exhibited a gradual weight loss throughout this second region, indicating that the decomposition of the ABS/CFRP matrix was a multi-step process rather than a single event. The temperature at which the maximum degradation rate occurred is referred to as the peak decomposition temperature (T_{max}).

By analysing the TGA curve, we can determine the thermal stability of the composite material. The onset temperature of degradation (T_{onset}) represents the temperature at which the weight loss starts, while the temperature at which 50% of the sample has decomposed (T_{50}) is often used to compare the thermal stability of different materials.

Additionally, the residual mass at the end of the TGA experiment provides insights into the char formation ability of the composite material. A higher residual mass indicates a better char formation, which can be beneficial in terms of fire resistance.

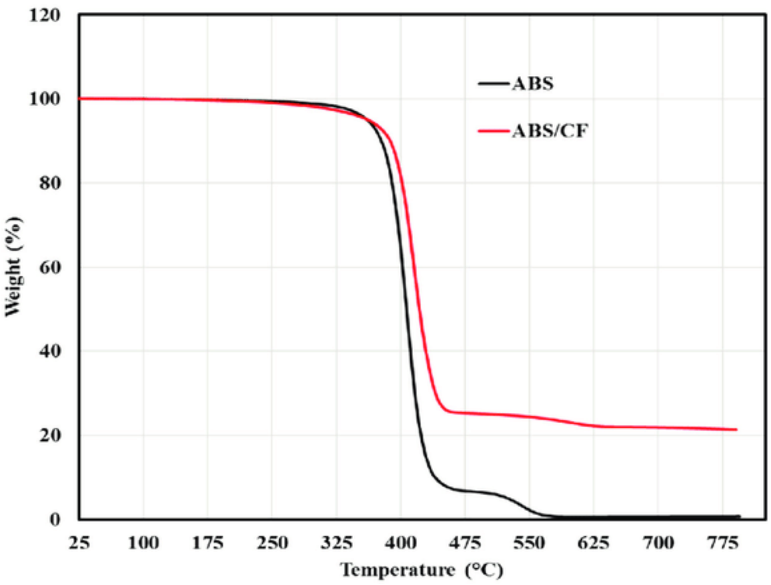

Figure 35 Thermogravimetric analysis of neat ABS and CF reinforced materials taken from [34].

Figure 35 shows the TGA curve of ABS and ABS with 20 wt. % CF. In case of the neat ABS, degradation occurs in two steps; fibre reinforced ABS also exhibit this behaviour i.e. where two major stages of weight loss are observed. The first stage started in the range of 300 °C and ended at about 450 °C, corresponding to the structural decomposition of the ABS polymer. The second stage started around 450 °C and ended at around 480 °C, which indicates the combustion of residual material.

The introduction of chopped CF into the ABS material reduced the thermal stability. To determine the thermal stability of the composite, decomposition onset temperature (DOT, defined as the 1% reduction of the weight) was identified from each test. In the case of 20 wt. % CF, DOT was found at 253 °C while the neat ABS had a DOT of 323 °C. Mixing chopped carbon fiber with ABS matrixes increased the thermal mobility of the composite material. As CF was introduced into the neat ABS, the reduction of DOT implied that the thermal energy dissipated by the composite has a path to conduct. Carbon fiber is superior to ABS in its ability to conduct thermal energy (thermal conductivity of CF is 1800W/mK while the ABS has 0.18 W/mK) thus the thermal gradient within matrix and fiber becomes higher, making the composite thermally unstable. Moreover, the thermal gradient within the composite helps to extrude the specimen at a relatively low temperature and extrusion force [34]

4.5. CAPILLARY RHEOMETRY

The capillary rheometry experiment of ABS/CFRP composite aimed to investigate the flow behaviour and rheological properties of the composite material. Capillary rheometry is a widely used technique to study the flow properties of polymers and their composites under different processing conditions.

The flow behaviour of the ABS/CFRP composite was evaluated by measuring the shear viscosity as a function of shear rate. Shear stress represents the force exerted by the material per unit area, while shear rate describes the velocity gradient within the material. This relationship between shear stress and shear rate is commonly known as the flow curve or viscosity curve. As can be seen in Figure 36, two tests were performed in the same material at 190°C. Similar shear-viscosity results were obtained for each material in the two tests made at the same temperature.

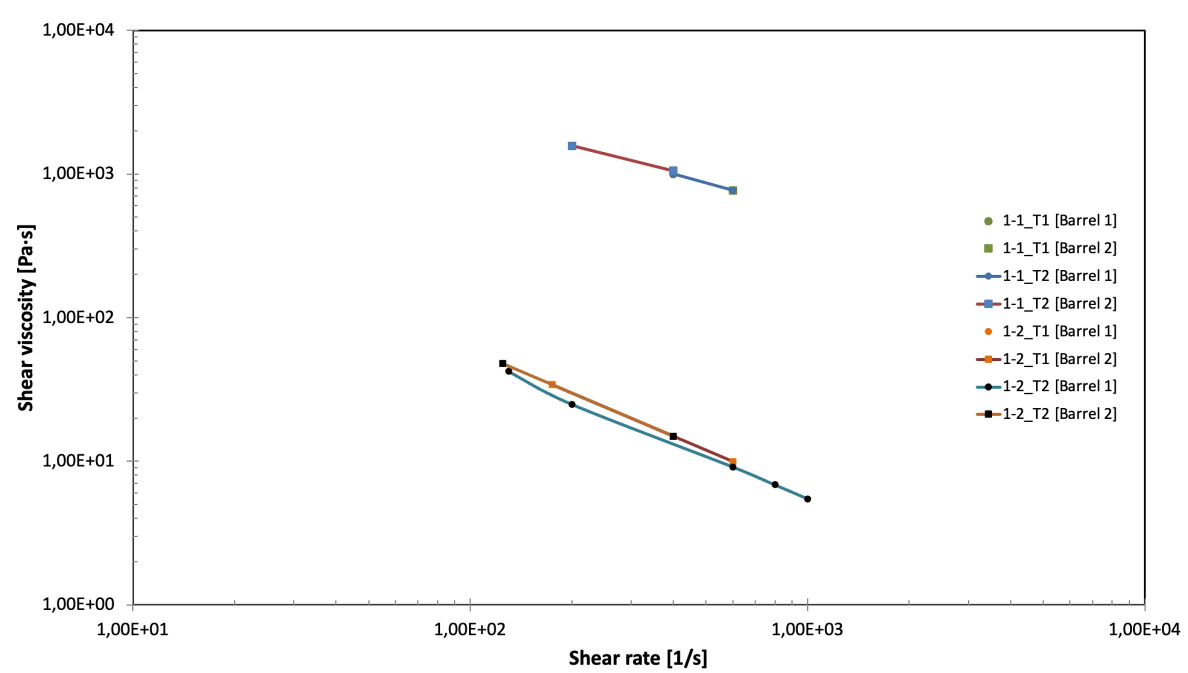

Figure 36 Viscosity Curve

The flow curve of the ABS/CFRP composite exhibited a non-Newtonian behaviour, which means that the viscosity of the material varied with the shear rate. At low shear rates, the composite displayed a shear-thinning behaviour, where the viscosity decreased as the shear rate increased. This behaviour can be attributed to the alignment and easier flow of the carbon fibres within the matrix at higher shear rates.

The presence of carbon fibre reinforcement in the ABS matrix influenced the rheological properties of the composite. Furthermore, the capillary rheometry experiment provided insights into the melt flow index (MFI) of the ABS/carbon fibre composite. MFI is a measure of the flowability of a molten polymer and is often used to assess the processability of materials. The MFI of the ABS/CFRP composite was found to be lower compared to pure ABS, indicating reduced flowability due to the presence of carbon fibre reinforcement.

The fibre volume influences viscosity. The composition with more fibre volume (1:1 ABS/CFRP) tends to have high viscosity compared to the material with more ABS. As expected, a decrease in carbon fibre decreases viscosity and increases flow properties

5. CONCLUSION

Thermoplastic matrix composites have gained significant attention in recent years due to their potential for enhancing the circularity of composite materials in various industries. The circularity of a material refers to its ability to be recycled, reused, or recovered at the end of its lifecycle, thereby reducing waste, and minimizing the depletion of natural resources.

Thermoplastic matrix composites offer several advantages in terms of circularity compared to other composites, such as thermoset matrix composites. One key advantage is their thermoplastic nature, which allows for the reprocessing and remoulding of the material multiple times, without significant degradation in mechanical properties. This means that at the end of their lifecycle, thermoplastic matrix composites can be melted down and reformed into new parts or products, enabling closed-loop recycling or upcycling processes.

The thermoplastic resin ELIUM is suitable for the resin infusion process and can be cured at room temperature, with the major advantages of being post-thermoformable, recyclable, and offering new possibilities for composite/composite or composite/metal assemblies. Good mechanical properties can be obtained using recycled CFRP after grinding through compression and extrusion. However, it is essential to carefully control the fibre volume fraction to avoid any degradation because it can increase the viscosity and resin impregnation process affecting, consequently, the composite’s mechanical properties.

The circularity of thermoplastic matrix composites makes them suitable for various applications across different industries. For example, in the automotive sector, these composites can be used for lightweight vehicles, reducing fuel consumption, and enhancing energy efficiency. At the end of a vehicle's life, the thermoplastic composites can be recycled and used in the manufacturing of new components, promoting a circular economy. Also, the reparability capacities of thermoplastic composites contribute to the increase of component’s service life.

In this work, long/short fibre materials, obtained from recycling continuous fibre composites, were demonstrated to be effective in reinforcing strengthening and toughening thermoplastic materials. It is reported that even small amounts of fibres drastically increased composite strength. It was possible to obtain reinforced materials from continuous fibre recyclates, showing outstanding material properties even when mixed with different thermoplastic matrixes, such as ABS. However, for high fibre volume, the dispersion and interface adhesion can be quite poor reaching lower stiffness and strength composite materials.

In conclusion, the study demonstrates that recycling continuous fibre thermoplastic composites is not only viable but also presents opportunities for applications in the same field as products made from virgin materials, leading to significant cost and environmental impact reductions. Conventional composite processing techniques such as VARI can finally be used for processing liquid thermoplastic resins and it can be concluded that thermoplastic matrix composites are a valuable alternative for the production of critical composite components.

6. SUGGESTIONS FOR FUTURE WORK

As the study has that it is possible to recycle post-industrial continuous fibre-reinforced thermoplastic composites.

The dependence of the mechanical behaviour on temperature, fibre volume and fibre length should be exhaustively evaluated to enable a good characterization of the material for structural analysis via the Finite Element Method. This will enable the structural optimization of components produced by the recycled composite.

To allow the production of geometrically more complex parts, a rheological study on the recycled composite should be carried out to enable the optimization of the injection moulding process for this typology of recycled materials. This should be supported through the definition of the composite recycled material rheological behaviour in the material database of an injection moulding numerical simulation software, which will enable the numerical simulation of the injection moulding process for the recycled composite material. Additional research is needed to study and find a better route for recycling thermoplastic composites the following are the suggestions.

- Involve process simulation for optimising and understanding a broad range of aspects associated with the process, such as mould design, fibre orientation, warpage, weld line prediction and the effect of process parameters.
- Develop more applications to increase knowledge and awareness of recycling thermoplastic composites.
- Test the applicability of design guidelines and knowledge of comparable processes, such as GMT and LFT.
- Update the LCCA with measured data and involve more environmental indicators.
- Study how to treat end-of-life waste and waste including impurities and investigate the effect on the application.
- Collaborate with partners in the supply chain, research and governmental institutes to obtain standardisation, preserve the traceability of materials and increase the motivation for recycling TPC.

7.REFERENCES

[1] Nagrale Priya, ‘Thermoplastic Composites Market’, *Market Research Future*, p. 140, Mar. 2023, Accessed: Aug. 04, 2023. [Online]. Available: https://www.marketresearchfuture.com/reports/thermoplastic-composites-market-4244

[2] A. Karakoç, V. K. Rastogi, T. Isoaho, B. Tardy, J. Paltakari, and O. J. Rojas, ‘Comparative Screening of the Structural and Thermomechanical Properties of FDM Filaments Comprising Thermoplastics Loaded with Cellulose, Carbon and Glass Fibers’, *Materials*, vol. 13, no. 2, p. 422, Jan. 2020, doi: 10.3390/ma13020422.

[3] N. L. Feng, S. DharMalingam, K. A. Zakaria, and M. Z. Selamat, ‘Investigation on the fatigue life characteristic of kenaf/glass woven-ply reinforced metal sandwich materials’, *Journal of Sandwich Structures & Materials*, vol. 21, no. 7, pp. 2440–2455, Oct. 2019, doi: 10.1177/1099636217729910.

[4] J. M. Winne, L. Leibler, and F. E. Du Prez, ‘Dynamic covalent chemistry in polymer networks: a mechanistic perspective’, *Polym Chem*, vol. 10, no. 45, pp. 6091–6108, 2019, doi: 10.1039/C9PY01260E.

[5] M. Bodaghi, C. H. Park, and P. Krawczak, ‘Reactive Processing of Acrylic-Based Thermoplastic Composites: A Mini-Review’, *Front Mater*, vol. 9, Jun. 2022, doi: 10.3389/fmats.2022.931338.

[6] W. Obande, C. M. Ó Brádaigh, and D. Ray, ‘Continuous fibre-reinforced thermoplastic acrylic-matrix composites prepared by liquid resin infusion – A review’, *Compos B Eng*, vol. 215, p. 108771, Jun. 2021, doi: 10.1016/j.compositesb.2021.108771.

[7] D. Bank, P. Cate, and M. Shoemaker, ‘pCBT: A New Material for High Performance Composites in Automotive Applications’, Oct. 2004. doi: 10.4271/2004-01-2698.

[8] Y. Qin, J. Summerscales, J. Graham-Jones, M. Meng, and R. Pemberton, ‘Monomer Selection for In Situ Polymerization Infusion Manufacture of Natural-Fiber Reinforced Thermoplastic-Matrix Marine Composites’, *Polymers (Basel)*, vol. 12, no. 12, p. 2928, Dec. 2020, doi: 10.3390/polym12122928.

[9] J. Zhang, V. S. Chevali, H. Wang, and C.-H. Wang, ‘Current status of carbon fibre and carbon fibre composites recycling’, *Compos B Eng*, vol. 193, p. 108053, Jul. 2020, doi: 10.1016/j.compositesb.2020.108053.

[10] S. J. Pickering, ‘Recycling technologies for thermoset composite materials—current status’, *Compos Part A Appl Sci Manuf*, vol. 37, no. 8, pp. 1206–1215, Aug. 2006, doi: 10.1016/j.compositesa.2005.05.030.

[11] G. A. Vincent, *Recycling of thermoplastic composite laminates : the role of processing*.

[12] C. Martin, ‘Wind turbine blades can’t be recycled, so they’re piling up in landfills’, Feb. 2020.

[13] G. Zhou *et al.*, ‘Experimental investigation on the effects of fabric architectures on mechanical and damage behaviors of carbon/epoxy woven composites’, *Compos Struct*, vol. 257, p. 113366, Feb. 2021, doi: 10.1016/j.compstruct.2020.113366.

[14] A. Shrestha, Y. Sumiya, K. Okazawa, T. Uwabe, and K. Yoshizawa, ‘Molecular Understanding of Adhesion of Epoxy Resin to Graphene and Graphene Oxide Surfaces in Terms of Orbital Interactions’, *Langmuir*, vol. 39, no. 15, pp. 5514–5526, Apr. 2023, doi: 10.1021/acs.langmuir.3c00262.

[15] ‘Carbon Fiber and Carbon Fiber Reinforced Plastic Market’, MarketsandMarkets.

[16] R. Dell’Anna, F. Lionetto, F. Montagna, and A. Maffezzoli, ‘Lay-Up and Consolidation of a Composite Pipe by In Situ Ultrasonic Welding of a Thermoplastic Matrix Composite Tape’, *Materials*, vol. 11, no. 5, p. 786, May 2018, doi: 10.3390/ma11050786.

[17] V. Sessini, A. J. López Galisteo, A. Leonés, A. Ureña, and L. Peponi, ‘Sandwich-Type Composites Based on Smart Ionomeric Polymer and Electrospun Microfibers’, *Front Mater*, vol. 6, Dec. 2019, doi: 10.3389/fmats.2019.00301.

[18] A. Zoller, P. Escalé, and P. Gérard, ‘Pultrusion of Bendable Continuous Fibers Reinforced Composites With Reactive Acrylic Thermoplastic ELIUM® Resin’, *Front Mater*, vol. 6, Dec. 2019, doi: 10.3389/fmats.2019.00290.

[19] M. I. Abdul Rasheed, ‘Compression molding of chopped woven thermoplastic composite flakes: a study on processing and performance’, University of Twente, Enschede, 2016.

[20] Thomas Alwart de Bruijn, ‘Recycling of Continuous Fibre Reinforced Thermoplastic Composites’, University of Minho, Guimaraes, 2020.

[21] A. M. Almushaikeh *et al.*, ‘Manufacturing of carbon fiber reinforced thermoplastics and its recovery of carbon fiber: A review’, *Polym Test*, vol. 122, p. 108029, May 2023, doi: 10.1016/j.polymertesting.2023.108029.

[22] M. Ö. Seydibeyoğlu *et al.*, ‘Review on Hybrid Reinforced Polymer Matrix Composites with Nanocellulose, Nanomaterials, and Other Fibers’, *Polymers (Basel)*, vol. 15, no. 4, p. 984, Feb. 2023, doi: 10.3390/polym15040984.

[23] Ē. Teirumnieka, K. Pīgožnis, D. Blumberga, E. Teirumnieks, and L. Lazov, ‘Processing composite materials with lasers’, *J Phys Conf Ser*, vol. 2487, no. 1, p. 012004, May 2023, doi: 10.1088/1742-6596/2487/1/012004.

[24] A. V. Muley, S. Aravindan, and I. P. Singh, ‘Nano and hybrid aluminum based metal matrix composites: an overview’, *Manuf Rev (Les Ulis)*, vol. 2, p. 15, Aug. 2015, doi: 10.1051/mfreview/2015018.

[25] T. Özben and N. Arslan, ‘FEM analysis of laminated composite plate with rectangular hole and various elastic modulus under transverse loads’, *Appl Math Model*, vol. 34, no. 7, pp. 1746–1762, Jul. 2010, doi: 10.1016/j.apm.2009.09.020.

[26] T. Ren, G. Zhu, and X. Hou, ‘The improved interface performance between carbon fiber and poly(ether-ether-ketone) by sulfonated polyether sulfone (<scp>s-PSF</scp>) sizing agent with different sulfonation degree’, *J Appl Polym Sci*, vol. 138, no. 19, May 2021, doi: 10.1002/app.50363.

[27] G. D’Emilia, A. Gaspari, E. Natale, and D. Ubaldi, ‘Uncertainty Evaluation in Vision-Based Techniques for the Surface Analysis of Composite Material Components’, *Sensors*, vol. 21, no. 14, p. 4875, Jul. 2021, doi: 10.3390/s21144875.

[28] L. H. C. Damacena, E. H. C. Ferreira, H. Ribeiro, G. J. M. Fechine, and A. M. C. Souza, ‘High-performance hierarchical composites based on polyamide 6, carbon fiber and graphene oxide’, *Polym Compos*, vol. 44, no. 6, pp. 3387–3400, Jun. 2023, doi: 10.1002/pc.27328.

[29] M. Topcu, E. S. Conkur, and G. Altan, ‘Elastic-Plastic Stress Analysis of Thermoplastic Composite Beams Loaded by a Single Force’, *Journal of Thermoplastic Composite Materials*, vol. 18, no. 3, pp. 181–193, May 2005, doi: 10.1177/0892705705043725.

[30] K. Chen, M. Jia, H. Sun, and P. Xue, ‘Thermoplastic Reaction Injection Pultrusion for Continuous Glass Fiber-Reinforced Polyamide-6 Composites’, *Materials*, vol. 12, no. 3, p. 463, Feb. 2019, doi: 10.3390/ma12030463.

[31] H. Tan *et al.*, ‘Crystallization and mechanical properties of carbon nanotube/continuous carbon fiber/metallocene polypropylene composites’, *Mater Res Express*, vol. 9, no. 1, p. 015302, Jan. 2022, doi: 10.1088/2053-1591/ac46e6.

[32] W. Oh, H. Lim, J. Won, and S. Lee, ‘Preparation of PVDF/PAR Composites with Piezoelectric Properties by Post-Treatment’, *Polymers (Basel)*, vol. 10, no. 12, p. 1333, Dec. 2018, doi: 10.3390/polym10121333.

[33] K. Verma *et al.*, ‘Development of Vacuum Enhanced Resin Infusion Technology (VERITy) Process for Manufacturing of Primary Aircraft Structures’, 2013. [Online]. Available: https://www.researchgate.net/publication/283495958

[34] K. M. M. Billah *et al.*, ‘Thermal Analysis of Thermoplastic Materials Filled with Chopped Fiber for Large Area 3D Printing’. [Online]. Available: https://www.researchgate.net/publication/341278269