



Review

Review on recycling of carbon fibre reinforced thermoplastics with a focus on polyetheretherketone

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ABSTRACT

Addressing the waste management of carbon fibre-reinforced thermoplastics, we review the different recycling routes, emphasising the mechanical recycling route of carbon fibre-reinforced polyetheretherketone (CF-PEEK). The most promising scheme for CF-PEEK is mechanical comminution, followed by long fibre-reinforced thermoplastics compression moulding. The main reasons are cost-efficiency and low environmental impacts, as it preserves the valuable matrix while enabling good mechanical properties. In this paper, we discuss the mechanical recycling route in general and then focus on the compression moulding step. Furthermore, we explore the effect on the mechanical properties to gain insights into potential fields of application for the mechanically recycled CF-PEEK. We also review the effect of CF-PEEK chemical degradation arising during compression moulding on the overall properties of the recycle. Understanding the mechanisms and changes in the fibre, matrix and fibre-matrix interface during recycling is crucial for optimising the process and maximising the number of recycling cycles.

1. Introduction

From 2011 to 2021, the European production volume of carbon fibre-reinforced plastics (CFRP) increased from 19,000 to 52,000 tons, with above-average growth of carbon fibre-reinforced thermoplastics (CFRTP) [1]. The waste management of CFRP is a crucial and evolving aspect of environmental sustainability and resource management. CFRP waste includes scraps, end-of-life products and manufacturing waste. Managing it reduces environmental impact and promotes circular economy principles. Two primary methods are currently in use. The first is landfilling, the prevailing method for industrial waste treatment, which will no longer be a viable option due to changing legislation. The second method is incineration, which has the advantage of using chemical energy from the waste by converting it to heat and electricity. However, carbon fibres are unsuitable for incineration plants since they do not thoroughly burn at the typical parameters of these plants. Hazardous fibres with a length of $>5 \mu\text{m}$, a diameter of $<3 \mu\text{m}$ and a length-to-diameter ratio of more than 3:1, according to the World Health Organization, can occur, which are significant health risks to humans since they can penetrate the deep airways [2]. The increasing demand

for lightweight materials [1] and the growing emphasis on environmental protection led to a significant interest in recycling. Recycling CFRP materials reduces waste and conserves the energy and resources that go into producing carbon fibres and polymer matrices. CFRP recycling's importance will likely grow as environmental concerns and regulations evolve. There are various recycling opportunities for CFRP, including thermal, chemical and mechanical recycling routes [2]. The choice of recycling method depends on several factors, such as the type of CFRP waste, the desired quality of the recycled material, economic feasibility, and environmental considerations.

This review focuses on CFRTP, the thermoplastic CFRP type commonly known as recyclable material due to the remeltability of the thermoplastic matrix [3]. However, a closer look at the recycling of CFRTP reveals that technological or economic challenges have stood in the way of industrial implementation. This problem is addressed in the following sections, taking the high-performance CF-PEEK as an example. Due to its resistance to high temperatures, chemicals and mechanical stress, CF-PEEK is gaining relevance for applications in hydrogen storage [4], aerospace, biomedical and gas and oil pipeline industries [5–8]. CF-PEEK generates significant economic interest in

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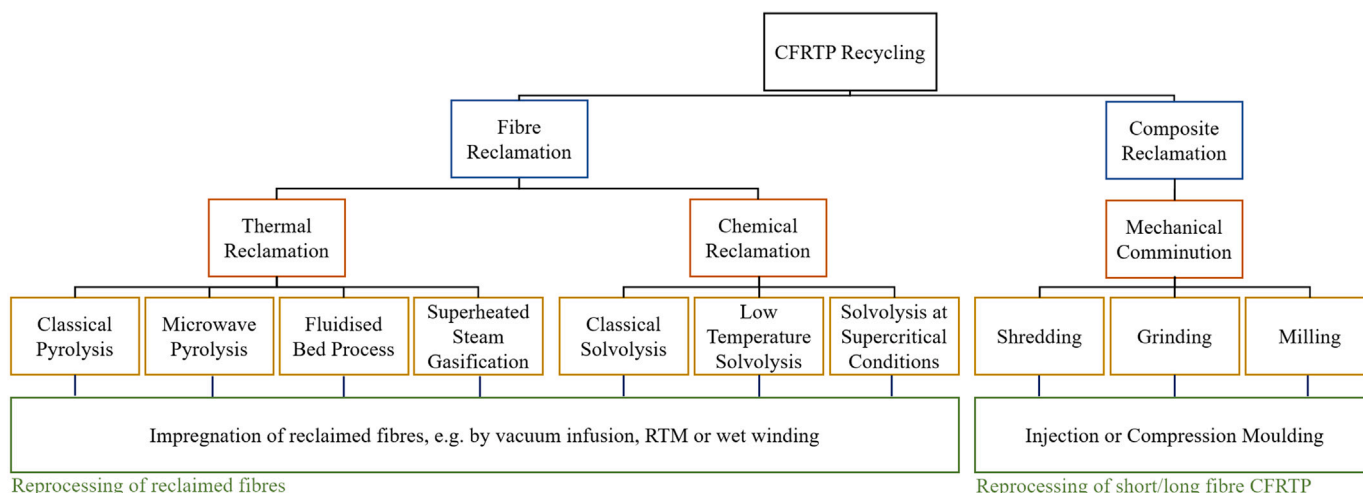


Fig. 1. Scheme of recycling methods for CFRTP.

being recycled as a whole composite without losing the high-quality matrix. However, comminution and reprocessing of CF-PEEK pose specific technological challenges, which is why CF-PEEK has not been mechanically recycled on an industrial scale to date. These challenges need to be addressed by research to make mechanical recycling of CFRTP possible, including high-performance composites such as CF-PEEK.

The paper aims to provide a comprehensive overview of the current state of the CF-PEEK mechanical recycling scheme through a comprehensive analysis of existing literature and case studies. To the authors' knowledge, no other reviews focus on the CF-PEEK mechanical recycling route, highlighting the potential benefits, limitations, and research gaps. This paper is a valuable resource for researchers, industry professionals, and policymakers interested in advancing sustainable practices and maximising the value of recycled materials.

2. Recycling methods for CFRTP

This section provides a comprehensive overview of possible approaches for recycling CFRTP, with a particular focus on CF-PEEK. As the data availability for CF-PEEK is partially incomplete, solutions for other CFRTPs are also presented, assuming transferability to CF-PEEK. One of the main objectives of recycling is to improve the environmental impact of materials. Section 2.5 summarises the data available for the LCA of the various recycling methods.

Two main approaches are commonly employed in the recycling of CFRTP. The first approach involves the separation of fibres from the matrix, subsequently processing these reclaimed fibres into new products. This separation is achieved through thermal or chemical processes that remove the matrix from the fibres. The second approach involves the reuse of the composite as a whole without separating the fibres from the matrix [9,10]. In this case, the waste material undergoes size reduction, melting, and reshaping to create a new part. Fig. 1 illustrates the various recycling routes available for CFRTP.

2.1. Thermal reclamation

Thermal reclamation methods are employed to recover the carbon fibres from their matrix by thermally decomposing the polymer matrix and, in some cases, recovering it. These processes take advantage of the distinct difference in thermal stability between carbon fibres and polymer matrices. Various approaches exist for heating the composite material and optional mechanical aids, which will be discussed in the subsequent sections.

2.1.1. Classical pyrolysis

In the context of recycling, pyrolysis refers to processes in which the composite waste is exposed to high temperatures ranging from 300 to 700 °C under a non-oxidative atmosphere. Under these conditions, the polymer matrix undergoes decomposition, producing pyrolysis gases such as CO, CH₄, and H₂, as well as pyrolysis oil, while the fibres can remain intact. To ensure the complete removal of any residues, an afterburning treatment with small amounts of oxygen or chemical treatment may be necessary [11]. By precisely controlling process parameters such as temperature and atmosphere pressure to match the characteristics of the recycled material, mechanical properties comparable to those of virgin fibres can be achieved [12]. This finding must be supplemented by the fact that the fibre properties can only be maintained if the post-industrial or end-of-life CFRTP component does not require mechanical shredding during preparation for pyrolysis. The energy requirements for pyrolysis-based waste processing have been reported to range from 3 to 48 MJ/kg depending on process variations [13,14].

2.1.2. Microwave pyrolysis

Microwave pyrolysis exploits the conductive properties of carbon fibres by inducing lossy eddy currents through high-frequency radiation, resulting in volumetric heating of the composite waste. Unlike conventional heating, microwave pyrolysis offers the advantage of significantly reducing the processing time, ranging from a few seconds to minutes, compared to approximately one hour in classical methods. Additionally, the controllability of the process parameters could be enhanced due to the fast adaptability of the radiation intensity. It is crucial to maintain an oxygen-poor atmosphere during microwave pyrolysis. Although microwave pyrolysis has a lower Technology Readiness Level (TRL) than classical pyrolysis, it has demonstrated promising outcomes in laboratory-scale experiments [15].

2.1.3. Fluidised bed process

The fluidised bed process (FBP) is a reclamation method that involves a highly accelerated stream of hot air at temperatures of 450–550 °C. This air stream is passed through a bed of quartz sand, with particles approximately 0.85 mm in size. After initial mechanical comminution, the composite waste is fed into this fluidised phase. As a result of the heat and mechanical impact of the fluidised phase, the polymer thermally decomposes. The fibres are carried along with the air stream as individual particles and are then captured. In the second stage, they can undergo post-treatment at around 1000 °C to

remove any remaining residues, similar to classical pyrolysis. Yip et al. conducted a study on the reclamation of CF-Epoxy Prepreg using the FBP process. They produced reclaimed carbon fibres (rCF) of up to 10 mm mean length with approximately 75% of their original tensile strength and similar to Young's modulus to the initial fibres [16]. In another study by Pickering et al. an industrial-scale fluidised bed reactor was constructed, which resulted in rCF exhibiting only an 18% loss of fibre strength and stiffness after recycling carbon fibre-reinforced epoxy waste [17]. It should be noted that the degradation of fibre properties primarily stems from sand particle abrasion [16]. Furthermore, the FBP process demonstrates excellent resistance to contaminants like metals [18]. Overall, the FBP offers a robust and efficient approach to recycling mixed carbon fibre-reinforced thermoset composites, with the potential transferability to CFRTP.

2.1.4. Supercritical steam gasification

A further technological approach, which can be assigned to the thermal recovery of CF, is the superheated or supercritical steam method. When heating water at a given pressure above the boiling point it will form dry steam used to fully degrade the polymer matrix of CFRP. A comprehensive review is provided by Hecker et al. [19], focusing on the most relevant research results on superheated steam gasification for reclaiming carbon fibres from epoxy [20–26] and, PA6 [27] based CFRP. Hecker et al. conclude that carbon fibres with a tensile modulus of 90%–100% and tensile strength of 65%–100% of the virgin CF properties can be reclaimed. The review states a technology readiness at laboratory or pilot-scale. Ongoing industry cooperations are investigating the method for a larger scale up to lower the energy use by up to 25% compared to using virgin fibres [19].

2.2. Chemical reclamation

Chemical reclamation processes constitute the second major group of methods for separating fibres and matrices in composite materials. These processes can be categorised into three main types: classical solvolysis, low-temperature solvolysis, and solvolysis at supercritical conditions.

2.2.1. Classical solvolysis

In classical solvent-based solvolysis, the polymer matrix is dissolved using polymer-specific solvents or solvent mixtures. This method allows for separating the dissolved polymer from the solution without depolymerisation. In some cases, the separated polymer can be purified and recovered, making it available for reuse through conventional plastic processing techniques. Solvent-based reclamation is still in the early stages of development for composite materials. However, significant progress has been made within the EU research project, "MultiCycle". The project focused on recycling CFRP waste with matrices such as polypropylene (PP), polyamide 6,6, polyamide 12, and bio-based polyamide. These matrices were derived from post-industrial automotive waste or end-of-life vehicle components [2]. Tapper et al. have demonstrated closed-loop recycling for CF-PP [28] and CF-PA6 [29] for fibres of 3 mm length. PP has been dissolved in xylene at 135 °C, PA6 in benzyl alcohol at 160 °C, both with a subsequent acetone treatment [28,29]. After compression moulding the reclaimed fibres and polymers showed no reduction in mechanical performance for CF-PP [28], whereas for CF-PA6 a decrease of approximately 40% in tensile properties was concluded [29]. Through the recycling process, these CFRP waste materials were successfully transformed into new composite materials, demonstrating the feasibility and potential of solvent-based reclamation for composite waste management.

A variation of solvent-based solvolysis is the solvent-based lamina separation, designed to separate only the lamina layers. Liu et al. proposed solvent treatment as a preliminary step in fibre reclamation. Rather than dissolving the entire thermoplastic surrounding each fibre,

they aim to selectively transfer only the material between fibre bundles to the solvent. In the case of Liu et al. carbon fibre-reinforced Polyetherimide (CF-PEI) is first cut into pieces and then immersed in a solvent bath, specifically N-methyl-2-pyrrolidone. Subsequently, the separated CF-PEI layers are transformed into single-layer chips, which are evenly arranged into the desired shape, such as a plate geometry using a vibrating plate. The thermoplastic solution obtained from the previous step undergoes filtration, followed by adding fresh polymer at 60 °C to ensure purity. The solution is then introduced into an airtight container with carbon fibre chips to prevent premature solvent evaporation. Once the fibres are thoroughly impregnated, the composite material undergoes heating above the solvent's boiling point (202 °C) to facilitate evaporation. Finally, the material is subjected to heat and pressure during compression moulding [30]. The disadvantage of this approach is the specific requirements for the solvolysis process corresponding to the laminate composition. Moreover, applying this approach to the more chemically resistant CF-PEEK would require the selection of a suitable solvent, such as sulfuric acid [31], which poses health risks [14]. An advantage over classical solvolysis could be preserving the fibre-matrix bonding, leading to better mechanical properties retention. Compared to mechanical shredding, this approach has the advantage that fibre lengths of already shortened fibres are retained in subsequent recycling stages.

2.2.2. Low temperature solvolysis

In solvolysis at low temperatures, the polymer matrix's chemical bonds are broken using reactive solvents. This process occurs under thermodynamically subcritical conditions, ensuring gentle treatment of the carbon fibres and preserving their properties. The monomers extracted from the solvolysis can be polymerised again for future use. Knappich et al. successfully recycled CF-reinforced Polyamide 6, Polyurethane and Epoxy composites using a multi-stage solvent extraction approach, maintaining the length and tensile strength of the recovered carbon fibres [32]. Furthermore, Buggy et al. demonstrated the complete dissolution of the matrix in CF-PEEK composites by utilising sulfuric acid at a temperature of 65 °C for 320 min [31]. However, it is essential to note that some solvents employed in solvolysis processes may risk human health and the environment [14].

2.2.3. Solvolysis at supercritical conditions

Supercritical Solvolysis closely resembles solvolysis at low temperatures, with the key difference being that the solvents are used in a supercritical state. When a fluid is heated and pressurised beyond its critical point, it exhibits properties between a liquid and a gas. Density and diffusion coefficients lie between liquids and gases, while viscosity and solubility resemble those of gases. Common solvents used in supercritical solvolysis include water, alcohol, and acetone [18]. Water, in particular, offers advantages such as ease of handling, non-toxicity, and recyclability. For example, in the case of an epoxy resin matrix, supercritical solvolysis requires conditions of 374 °C and 22.1 MPa, resulting in a matrix removal efficiency of 99.5% [33]. Alternatively, certain alcohols can be used at lower temperatures and pressures, such as 200–300 °C and 2.0–6.0 MPa for epoxy resin [34]. Adding a catalyst, possibly in combination with oxygen, can enhance the effectiveness of the process and facilitate the attainment of the desired process parameters [35,36].

Dandy et al. successfully applied supercritical solvolysis to CF-PEEK using a mixture of 20 vol-% ethanol and 80 vol-% water, catalysed by caesium carbonate Cs_2CO_3 at 350 °C and 16.8 MPa for 30 min, resulting in clean carbon fibres [37]. However, it is essential to note that the process can be relatively expensive due to the complex reactor technology required [12]. Thus, further research and development are necessary to optimise process parameters, catalyst selection, and reactor design to enhance the cost-effectiveness and scalability of supercritical solvolysis methods.

2.3. Mechanical comminution

Mechanical comminution comprises processes in which composite materials are recovered as a whole composite. This distinguishes the processes from those in which the polymer matrix and fibres are separated first, such as in thermal and chemical reclamation methods [38, 39]. Thanks to this direct process chain, the mechanical recycling scheme is usually an environmentally friendly and energy-efficient method. Mechanical comminution generally applies to all types of fibre-reinforced plastics and involves several stages of size reduction of composite waste. Coarse crushing or shredding can be a preparation for the recovery of long fibres using thermal or chemical reclamation methods for thermoset or thermoplastic composites. In the case of CFRTP, size reduction is often aimed at direct reprocessing of the recyclate. The recyclate can be reprocessed as long-fibre or short-fibre reinforced CFRTP in downstream processes such as compression or injection [40,41].

2.3.1. Shredding

Various process chains emerge, offering options for size reduction. The predominant method involves shredding or crushing devices, which yield a diverse range of output materials in size and composition. Achieving a uniform output in particle size distribution is desirable but can prove challenging. Consequently, specific processes may necessitate classification methods such as sieving [2,42].

In general, the shredding process is accompanied by the shortening of fibres and a mild degradation of the matrix material due to cutting forces. However, it is essential to note that size reduction also brings benefits such as enhanced processability and improved manufacturing of complex geometries particularly through compression moulding [43,44]. Among the established methods for CFRTP size reduction, shredding stands out as an effective method that provides extensive options in terms of material output size. Unlike other methods like hammer-milling, rotary-cutting milling or electro-dynamical fragmentation, multiple-shaft shredding can produce larger and more uniform flakes. The key advantage of this shredding method lies in its principle of shearing the input material rather than subjecting it to impacts, making it well-suited for the mechanical comminution of high-performance CFRTP with high toughness and impact resistance, such as CF-PEEK [45]. Commonly used shredder models for CFRTP include 2-shaft or 4-shaft shredders, as schematically shown in Fig. 2.

During shredding, the waste material is fed into the shredder, where teeth on the rotating inner blades compel it to move downwards within the blue-hatched areas shown in Fig. 2. As the rotating blades overlap,

they shear the material in the orange hatched area. A screen is positioned directly beneath the blades, and depending on size, the resulting flakes will either pass through the screen or be captured by the teeth on the outer shafts and moved upwards for further shredding. There is no analytical formula to calculate the flakes' exact particle size distribution (PSD) based on shredding settings. The flakes' fibre length distribution (FLD) is primarily influenced by the laminate structure of the CFRTP part being recycled and secondarily by the PSD associated with the adjusted blade width and the screen size in the shredder [45]. While variations in rotational speed and blade sharpness significantly impact the fracture mechanism and the achievable feed rate, they have minimal effect on the resulting flake sizes [46]. Furthermore, varying the sizes of end-of-life (EoL) parts to be comminuted has shown no significant influence on the resulting FLD [45]. Fig. 3 illustrates representative FLD probability density functions of shredded multilayered CF-reinforced Polyphenylene sulfide (CF-PPS) scrap, demonstrating a prominent peak at the utilised blade width (in the absence of a screen) and a wide dispersion of fibre lengths beyond the standard deviation [45].

The distribution shown in Fig. 3 can be attributed to two effects during the shredding process. First, flakes may undergo multiple cuts, resulting in widths smaller than the blade width. Secondly, large fragments can be generated as by-products and pass through the shredder and the screen. To minimise the size deviation and achieve the desired FLD, thus the desired mechanical properties of the recycled part, it is crucial to determine the PSD and FLD of the sorted flakes [45].

The classification of shredded composite scrap can include two stages, depending on the requirements. In the first stage, screening methods classify flakes based on their PSD. In the second stage, flakes can be analysed according to their FLD. When dealing with unknown CFRTP waste, the fibre volume content (FVC) can be a further important parameter to be analysed. With constant shredding parameters, such as constant blade width and material feed rate, the FLD can be determined once for a representative quantity of flakes and transferred to the entire batch [45]. To classify the flake size cost-effectively, using sieves, such as multi-stage vibrating sieves, proves effective. Multi-stage vibrating sieves contain perforated plates with round or square apertures. The configuration of these staged sieves and the impact of screen sizes and plate distances on the resulting PSD in each stage are standardised and specified in the ASTM E323 standard specification [48]. Another more elaborate method for classifying shredded CFRTP is a cascade air classifier, such as a zig-zag air classifier. This classification technique utilises controlled airflow, gravity, and material drag to separate materials of different shapes and sizes. Each classification stage produces two grades known as coarse and fine cuts. Different, repeatable settings can be established for each classification stage by

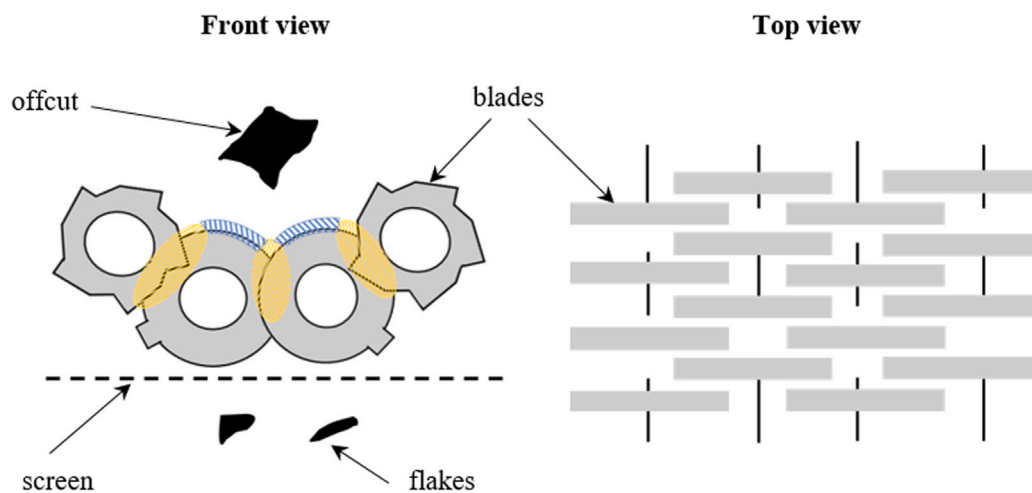


Fig. 2. Schematics of a 4-shaft shredder, front (left) and top view (right).
Source: Adapted from [45].

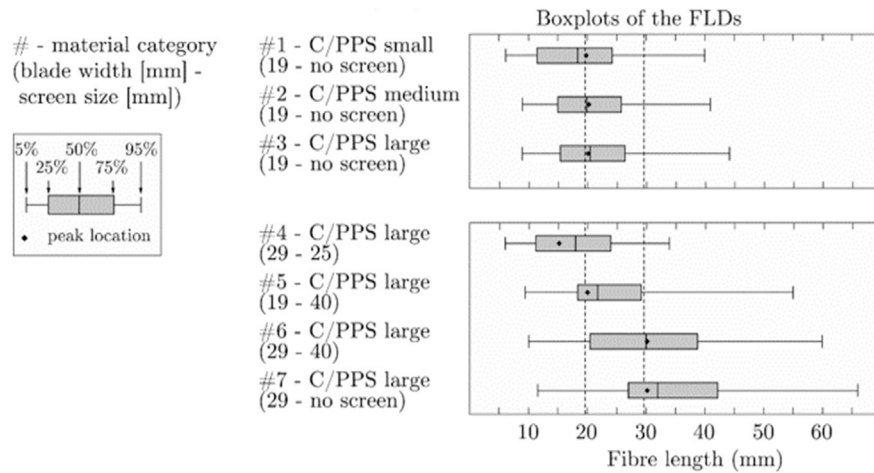


Fig. 3. Resulting FLD for different blade widths, screen sizes and sizes of the CF-PPS (C/PPS) input composite scrap. Source: Adapted from [47].

airflow through the system. An advantage of the air classifier method is the automatic de-dusting of the size-reduced material through the airflow and its ability to handle an extensive range of particle sizes [49]. The FLD is often estimated to be equal or strongly correlated with the classified flake size. However, this assumption may not always hold, particularly when dealing with multilayered flakes where fibre orientations vary. In that case, precise conclusions cannot be drawn based solely on available PSD information [45].

Several optical analysis methods, such as image processing or fibrographical methods, are being researched to determine the FLD during the sieving stage. However, in most optical analysis methods, the matrix of the composite part needs to be removed to enable FLD determination through optical analysis methods. The matrix removal is typically achieved through pyrolysis or fluidised bed treatment. Notably, these matrix removal techniques often involve high energy consumption and result in the shortening of the carbon fibres. As a result, the FLD measured via image analysis represents a conservative estimation of the actual FLD [50–52]. A more convenient method for determining the FLD without matrix destruction or fibre degradation is the measured orientation method introduced by Vincent et al. [45]. This method is applicable when the ply layup of the composite is known, such as in the case of in-house waste. To implement this method, a representative batch of flakes is sampled at a specific sieving stage. Photographs of the flakes are taken, and the fibre orientation of the top layer is determined using image analysis. The fibre orientations of each ply are then converted into arrays. In the final step, the binary images and their arrays are superimposed, and the FLD is determined by calculating the line length distribution of the intersected images [45].

2.3.2. Grinding and milling

Although grinding, milling and shredding are often used interchangeably, they are different processes with different results. In contrast to grinding and milling, shredding is a process characterised by the cutting, tearing or shearing material into larger, irregular fragments. While all the processes contribute to size reduction, the difference lies in the size and uniformity of the resulting particles. In grinding and milling, a material is carefully crushed to achieve a fine structure, with the end product consisting of smaller, more uniform particles. This precision is particularly valuable for injection moulding that requires consistent small particle sizes and fibre length distribution [40]. A comprehensive review on milled carbon fibres by Newman et al. discusses further areas of application and properties of milled carbon fibres. Considering a typical fibre length of $\sim 100 \mu\text{m}$ after milling virgin or recycled carbon fibres, potential applications include structural

composites, electrical conductivity, sensor technology and multifunctionalisation of materials. Most commonly used technologies are ball milling or hammer milling. In ball milling, the composite or carbon fibres are placed in a drum containing hardened balls with a high density. By rotating the drum, the balls collide with the composite or fibres and produce a fine powder through high-pressure impacts. The powder is then filtered through a sieve that can be adjusted to the desired powder coarseness. In hammer milling, a series of mounted hammers impact at high speed on a fixed screen bottom [53]. According to Li et al. hammermilling can also be used as an alternative to shredding to recover larger fractions of recycled composites. The study shows better mechanical properties for the compression moulded parts made from shredded recycle comparing to hammermilled counterparts, although without statistical significance [42].

2.4. Other recycling methods

2.4.1. Enzymatic decomposition

Enzymatic decomposition, developed at the Hohenstein Institute, aims to decompose the matrix polymer using biological processes without damaging fibres. Initial experiments show that the biological decomposition of epoxy resin could be possible [54,55]. Although in a very early stage of development, the technology is attractive in terms of its boundary conditions. It offers the potential to require little energy input, the reuse of decomposed matrix products, and no environmentally critical solvents. Even though this could be a highly interesting approach for CFRP in general, it does not offer any proven research successes at the present time. For the biocompatible and enzyme-resistant PEEK in particular, it is questionable whether enzymatic decomposition can be achieved in future research.

2.4.2. Electrodynamical fragmentation

Electrodynamical fragmentation (EDF) offers an alternative to shredding or crushing for breaking down CFRTP. In this method, composite waste is immersed in a water bath between two electrodes, which receive electrical pulses from 50–200 kV. The voltage applied must surpass the breakdown voltage of the solid waste material while remaining lower than that of water, which is achieved by employing rise times of less than 5 microseconds. The discharge results in a high energy density of approximately $10\text{--}100 \text{ J/cm}^2$, inducing a plasma channel in the solid. As a result, temperatures of up to $10,000 \text{ }^\circ\text{C}$ and pressures of 104 MPa are generated within a confined area, generating shock waves that surpass the matrix strength and induce cracks. Roux et al. demonstrated the functionality of the EDF process by comminuting hinges from a helicopter door and remanufacturing the same part using the recovered

CF-PEEK. Throughout six EDF cycles, which included a sieving step after each application, 60% of the initial material could be processed into uniformly sized fragments suitable for reuse. The resulting parts retained 83% of the original mechanical properties [80,81].

2.5. Assessment of the environmental impact

One of the primary goals for recycling is to reduce the environmental impact of a product system. To communicate that environmental impact of recycling methods, they are assessed as part of life cycle assessments (LCA), capturing material input and waste output throughout the whole life span of a product [82]. A comprehensive study on the evaluation of LCA and its application to closed-loop recycling of CFRP was carried out by Tapper et al. [83], which contains results cited below, among others. In this section, particular emphasis will be placed on the energy usage of the above-listed recycling methods, as it is often regarded as the most influential parameter when discussing environmental impacts.

With 183–771 MJ/kg [13,84–89], carbon fibres are often the most energy-intensive part of CFRTP components, giving particular reason to recover carbon fibres for several life cycles. In the case of high-performance thermoplastics such as PEEK, the matrix component also has a high energy-intensive production phase [90]. Borda et al. assume an energy consumption in the range of 283 to 298 MJ/kg to obtain PEEK matrix for CF-PEEK [91]. For the pyrolysis of CFRTP waste, the reported energy requirements range from 3 to 48 MJ/kg waste [13,14,64,65]. Simultaneously, the greenhouse gas potential is shown to be 2.9 to 5.9 kg CO₂-eq./kg waste [13,14]. Chemical reclamation, on the other hand, has an energy requirement of 7 to 19 MJ/kg waste [14,76], with a greenhouse gas potential of 1.2 to 1.5 kg CO₂-eq./kg waste [14,92]. Khalil et al. compared pyrolysis with supercritical water solvolysis and concluded that supercritical water solvolysis has a significantly 78 times higher human health impact, 76 times ecotoxicity, 17 times global warming potential, and three times ozone depletion potential compared to pyrolysis [93]. Keith et al. found an energy requirement of 19 MJ/kg reclaimed CF using a supercritical mixture of acetone and water [76]. Another study [77] showed an energy range of 60 to 90 MJ/kg for a process that utilises benzyl alcohol as a solvent

and tripotassium phosphate as a catalyst at 200 °C and atmospheric pressure. Meng et al. conducted several studies with the fluidised bed process (FBP). They demonstrated that recovering CF by FBP exhibits 33%–51% lower greenhouse gas potential and requires 32%–50% less primary energy consumption compared to production of virgin CF [94]. For the FBP, an energy consumption of 6 MJ/kg waste was documented by Meng et al. [13]. In non-separating recycling process schemes, which primarily involve the comminution step for CFRTP waste, shredding requires 2.0 MJ/kg at a throughput of 10 kg/h. At a higher rate of 150 kg/h, energy consumption could be reduced to 0.3 MJ/kg [78]. The energy requirement for compression moulding of shredded CFRTP is determined to be 10 MJ/kg by Suzuki and Takahashi et al. [95].

2.6. Conclusion on the recycling methods for CFRTP

Throughout this section, research findings on recycling methods for CFRP have been reviewed. Various technological approaches have been identified and analysed based on their capabilities and requirements regarding energy, as well as material input and output, as summarised in Table 1. Most methods can be categorised into thermal, chemical, or mechanical reclamation. Currently, no all-CFRP-encompassing solution makes the recycling of thermoplastic CFRP attractive for industrial applications. The focus of available research results lies in the thermal and chemical recovery of carbon fibres from thermoset composites. An important factor to consider is that also for thermal or chemical reclamation, most post-industrial or end-of-life waste must be mechanically comminuted in the first processing step. The crosslinked nature of thermosets subsequently necessitates the complete decomposition of the matrix, with few exceptions, to recover the valuable carbon fibres. The literature shows that significant process energy or potentially harmful solvents must be used for this polymer removal. In contrast, for thermoplastic composites, non-separating methods via mechanical comminution and direct reprocessing can be advantageous in terms of process costs, energy consumption, and environmental impact. This approach is particularly reasonable for CFRTP with high-value thermoplastics like PEEK that come with energy-intensive production and high-performance properties. Preferably, a process that allows the recycle to be reused with a high fibre volume content and long rather

Table 1
Recycling methods for CFRTP including processing inputs and outputs.

Process	Input	Output	Authors
Thermal reclamation			
Classical pyrolysis	<ul style="list-style-type: none"> Heat (300–700 °C) Non-oxidative atmosphere CF/epoxy, CF/Polybenzoxazine 3 to 48 MJ/kg 	<ul style="list-style-type: none"> Pyrolysis gases (e.g. CO, CO₂, CH₄, N₂, H₂, C₂H₆) Pyrolysis oils (e.g. benzene, toluene, ethyl benzene, phenols) Carbon fibres 	[11–14,41,56–65]
Microwave pyrolysis	<ul style="list-style-type: none"> High-frequency radiation (2.45 GHz) Process gas (Air, N₂ or Ar) CF/epoxy 	<ul style="list-style-type: none"> Pyrolysis gases Pyrolysis oils Carbon fibres 	[15,66–69]
Fluidised bed process	<ul style="list-style-type: none"> Hot air (450–550 °C) Quartz sand Comminuted composite waste (CF/Epoxy ≤ 25 mm) Post treatment heat (1000 °C) e.g. 6 MJ/kg 	<ul style="list-style-type: none"> Pyrolysis gases Short/long carbon fibres 	[13,16–18]
Supercritical steam gasification	<ul style="list-style-type: none"> CFRP (CF/Epoxy, CF/PA6) Superheated steam (500 °C to 800 °C) In some cases process gas N₂ or superheated CO₂ 	<ul style="list-style-type: none"> Carbon fibres Non-condensable gas fraction (low-molecular weight hydrocarbons such as CO, CO₂, CH₄, H₂) Gases to be condensed into oils (pyridines, benzenes, phenols) 	[11,19–27]

(continued on next page)

Table 1 (continued).

Chemical reclamation		
Classical solvolysis	<ul style="list-style-type: none"> • Polymer-specific solvents or solvent mixtures (e.g. water, alcohol, ammonia, glycol, nitric acid, phosphoric acid) • Heat (90 °C–280 °C) • Pressure (0.1–4 MPa) • CF/PP, CF/PA 6 CF/PA 6.6, CF/PA 12, CF/PEI, CF/Epoxy • e.g. 7.6 MJ/kg 	<ul style="list-style-type: none"> • Dissolved polymer • Solvents • Carbon Fibres <p>[2,14,18,28–30,70]</p>
Low temperature solvolysis	<ul style="list-style-type: none"> • Thermodynamically subcritical conditions (e.g. 65 °C) • Polymer-specific solvents or solvent mixtures (e.g. sulfuric acid, peroxide hydrogen, N,N-dimethylformamide, polyethylene glycol, dimethylformamide) • CF/PA6, CF/PU, CF/Epoxy, CF/PEEK 	<ul style="list-style-type: none"> • Dissolved polymer • Solvents • Carbon fibres <p>[31,32,71–75]</p>
Solvolysis at supercritical conditions	<ul style="list-style-type: none"> • Solvents in supercritical state (e.g. water, alcohol, acetone) • Heat (200–374 °C) • Pressure (2–22 MPa) • In some cases catalysts (e.g. caesium carbonate) and oxygen • CF/Epoxy, CF-PEEK • 19 to 90 MJ/kg 	<ul style="list-style-type: none"> • Dissolved polymer • Solvents • Carbon fibres <p>[18,33–37,76,77]</p>
Mechanical comminution		
Shredding	<ul style="list-style-type: none"> • Power supply for the shredder and sieving/classification stage • Continuous/long/short fibre-reinforced thermoset or thermoplastic Composites (e.g. CF/PEEK, CF/PPS) • 0.3 to 2.0 MJ/kg 	<ul style="list-style-type: none"> • Long/short fibre-reinforced composites • Larger and more uniform flakes compared to milling <p>[2,40–45,49,78]</p>
Grinding and milling	<ul style="list-style-type: none"> • Power supply • Comminuted composite waste • e.g. 2 MJ/kg 	<ul style="list-style-type: none"> • Short fibre-reinforced composites or powders • Smaller, more uniform particles <p>[40,53,79]</p>
Other recycling methods		
Enzymatic decomposition	<ul style="list-style-type: none"> • Microorganisms or enzyme producing Fungi • Little energy input • No solventsCF/Epoxy 	<ul style="list-style-type: none"> • Carbon fibre • Fermented/degraded epoxy residues (not yet clearly proven) <p>[54,55]</p>
Electrodynamic fragmentation	<ul style="list-style-type: none"> • Power supply for electrical pulses (50–200 kV) • Comminuted composite waste (e.g. CF/PEEK) 	<ul style="list-style-type: none"> • Long/short fibre-reinforced composite fragments <p>[80,81]</p>

than short fibres to achieve maximum mechanical properties should be selected. Consequently, the preferred recycling route for CF-PEEK involves mechanical comminution followed by compression moulding using long fibre-reinforced CF-PEEK recyclate.

3. Reprocessing of CF-PEEK recyclate via compression moulding

Alongside other carbon fibre-reinforced high-performance polymers such as CF-PEKK or CF-PAEK, CF-PEEK is one of the highest-quality CFRTPs used industrially. Its current applications are limited to niche areas in medical technology and aerospace. However, the demand for its use in aerospace may significantly increase due to the growing need for automation in manufacturing processes and the recyclability requirements, aiming to replace conventional thermoset CFRP. Fibre-reinforced thermoplastics are believed to have good recyclability due to the repeatable meltability of the thermoplastic matrix. The properties of recycled CF-PEEK and the parameters affecting these properties will be discussed below according to the state of the art. Particularly for high-performance CFRTP like CF-PEEK, there are gaps in the available data, so characteristics of comparable CFRTP will also be presented below.

3.1. Process chain for reprocessing of CF-PEEK recyclate via compression moulding

Due to the inherent challenges and significant environmental drawbacks associated with thermal and chemical reclamation methods, mechanical comminution has emerged as the predominant approach for CF-PEEK [96]. As shown in Fig. 4, the mechanical recycling scheme involves the collection of end-of-life (EoL) CFRTP scrap, followed by a series of shredding or crushing processes to reduce it into smaller pieces, commonly known as flakes or chips. Subsequently, the shredded waste is classified to achieve a controlled PSD and FLD before compression moulding [2].

After collecting CF-PEEK offcuts and EoL scrap, shredding can produce flakes ranging from 1 to 250 mm in diameter to meet the requirements for their use as reinforcement in recycled long-fibre thermoplastics (rLFT). As fibre length significantly influences the mechanical properties of CFRTP, the classification of shredded CF-PEEK into groups of different fibre length distribution (FLD) is an essential step, as described in Section 2.3.1. After shredding and classifying the composite waste, different processes can be applied to reshape the material to a desired geometry. Injection moulding is a viable process for recycling short fibre-reinforced thermoplastic (SFT) waste. The shredded and milled or ground SFT can be fed directly or compounded

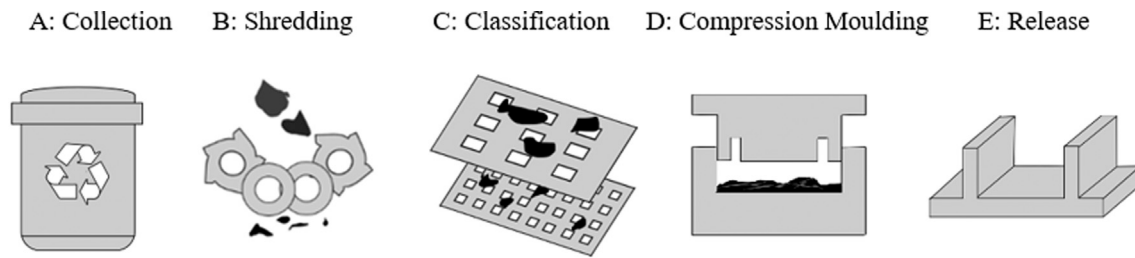


Fig. 4. Flow chart for compression moulding of CF-PEEK recycle.
Source: Adapted from [45].

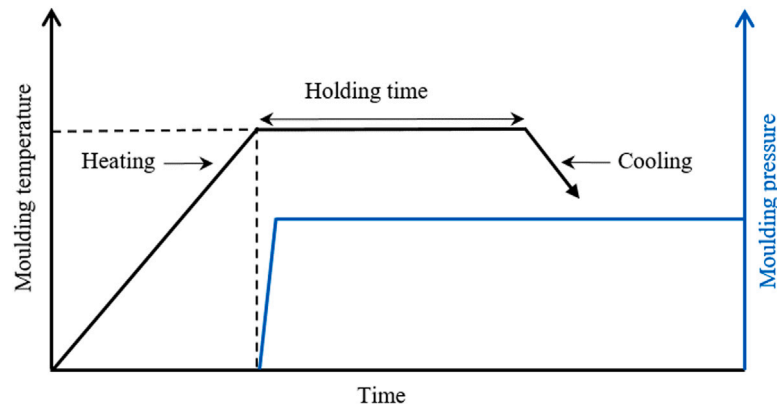


Fig. 5. Schematic cycle for CF-PEEK compression moulding with moulding temperature, heating rate, holding time, cooling rate and consolidation pressure.

into an injection mould. The fibre weight fraction is limited to approximately 50% to ensure good processability [97]. The mechanical properties of recycled SFT parts can reach around 90% of virgin injection moulded parts [98]. A suitable process for recycling CF-PEEK as long fibre-reinforced thermoplastic (LFT) is compression moulding.

Compression moulding involves heating CF-PEEK under high pressure, within a closed mould cavity until the matrix consolidates. The matrix undergoes liquefaction and flows during this process, conforming to the cavity geometry. Subsequently, it solidifies into the desired product as it cools. Once the part has sufficiently cooled, it is removed from the mould. The overall CF-PEEK compression moulding process and its main process parameters are illustrated in Fig. 5.

Compression moulding is suitable for a wide range of industrial and commercial consumer parts and products ranging from small to large automotive body parts. When compression moulding long fibre-reinforced thermoplastics, their flow properties allow more complex part geometries compared to continuous fibre-reinforced plastic parts.

Due to the local structural integrity of the chips, large chip sizes can cause blockages in the flow paths and damage to the tool. Larger chips result in higher strengths and larger scatter when measuring mechanical properties, making a reliable prediction of material behaviour difficult. In the case of variothermal processing, the compression moulding has comparably long cycle times caused by the tool's need to heat the material above its melting temperature and then cool it down again for demoulding. Compression moulding of long fibre-reinforced thermoplastics compromises the good mechanical properties of continuous fibre-reinforced plastics and the possible geometric complexity of short fibre-reinforced plastics [99]. The resulting mechanical properties are lower than continuously fibre-reinforced parts but significantly higher than those of short fibre-reinforced parts. Enhanced geometric complexity means greater design freedom with efficient functional integration and overcoming the typical limitations of shell-like structures in conventional laminate systems.

De Bruijn et al. proposed an optional advancement of LFT compression moulding, introducing an intermediate mixing step and simultaneously pre-melting in a low-shear mixing device [100]. The

mould can stay at a constant temperature with the LFT already molten before filling the cavity, reducing the cycle time to a few minutes. Additionally, the pre-mixing, done at low shear rates to prevent fibre damage, dissolves the laminate structure into centimetre-sized CFRTP chips after shredding. De Bruijn et al. have shown that mixing improves the mechanical properties compared to direct LFT compression moulding. Fibre accumulation can occur during mixing and lead to a limited fibre volume content, which can be a downside of low-shear mixing. Nevertheless, up to 50% of FVC materials have been successfully processed [45,92,100].

The impact of the fibre length and compression moulding parameters on the mechanical properties of high-performance CF-PEEK is examined in more detail in Section 3.2. Due to the above-mentioned gaps in available data for high-performance CFRTP, the properties of comparable CFRTP are also presented below.

3.2. Effects of compression moulding parameters on mechanical properties

3.2.1. Effects of the fibre length

Fibre length is critical for CFRP's strength and stiffness. The categorisation into short, long (also referred to as discontinuous), or continuous fibre-reinforced composites has to be done by the critical length of the fibre within a specific matrix. To give an idea of the order of magnitude of this categorisation, in the publication of Heim, for example, fibres shorter than 1 mm are considered short carbon fibres (SCF), those longer than 1 mm are considered long carbon fibres (LCF) and those longer than 50 mm are considered continuous fibres [101].

In available literature, the effect of flake size or fibre length distribution on mechanical properties is mostly examined using compression moulded randomly oriented strands (ROS). Selezneva compares compression moulded CF-PEEK ROS in a range of 3 mm to 12 mm strand width and 6 mm to 50 mm in strand or fibre length and finds an increase in in-plane tensile, compressive and shear strength and modulus for longer fibres. This improvement can be attributed to the overlapping and formation of a laminate-like layer structure when

large flakes align during ROS compression moulding. The flakes are compression moulded at 380 °C for 15 min with a consolidation pressure of 3.5 MPa and a 10 °C/min cooling rate. The microstructure exhibits a uniform laminate layer where the fibres are evenly aligned on top of each other, forming a cohesive structure without any notable defects. In contrast, looking at the microstructure of a ROS sample made from smaller flakes, stronger waviness can be seen. In this case, flake orientations deviate from a horizontal orientation in some regions. Due to this displacement out of the horizontal direction, areas with larger matrix content could occur. These irregularities result in lower tensile properties in these regions [102]. The work of Leger et al. support the increase in shear modulus and shear strength with longer fibres [103]. Some of their ROS specimens containing long strands exhibit higher shear properties than quasi-isotropic CF-PEEK laminates [103]. Li et al. compare CF-PEEK with 200 µm fibre length to 2–3 mm fibre length [104]. By increasing fibre length, a significant improvement in compressive strength from 196 MPa to 449 MPa is observed. Similarly, the tensile strength increases from 137 MPa to 388 MPa, and flexural strength from 259 MPa to 547 MPa [104]. McGrath et al. conducted a study investigating the relationship between flake sizes and resulting mechanical properties, including compressive strength, tensile strength and flexural strength [105]. The study utilised perfectly uniform squared flakes obtained from CF-PEEK prepreg with an FVC of 64%. The flakes were cut into various lengths ranging from 6.35 mm to 25.4 mm and were then processed through compression moulding at 390 °C, a consolidation pressure of 5.5 MPa and a cooling rate of 7 °C/min. Randomly scattering the flakes into the mould led to a random fibre orientation and approximately quasi-isotropic specimens. The results show a linear dependence between flake sizes and mechanical properties. As the fibre length increases from 6.35 mm to 25.4 mm, the flexural strength exhibits a linear increase from 370 MPa to 450 MPa, the compressive strength increases from 330 MPa to 440 MPa, and the tensile strength increases from 150 MPa to 230 MPa. The study further states that fracture always tends to follow the shortest path in a structure. Consequently, in a matrix reinforced with short fibres and thus a shorter overlap of fibres, the distance a crack has to overcome is shorter than with longer fibres. This results in a lower stress required for crack propagation, leading to a reduction in the mechanical properties of the part. Due to shorter fibres and random orientation of flakes, the mechanical properties are significantly lower compared to virgin CF-PEEK unidirectional $[0]_{16}$ laminates reaching 2125 MPa tensile strength, 1094 MPa in compressive strength and 1876 MPa flexural strength. Compared to the results for $[90]_{16}$ laminates with 140 MPa flexural strength and 80 MPa tensile strength, the ROS samples perform significantly better. The ROS samples are best compared with the quasi-isotropic (QI) $[+45, 90, -45, 0]_{2,5}$ layup. The tensile strength of QI at 704 MPa is significantly higher than that of ROS (25.4 mm) at 230 MPa. The flexural strength of QI at 616 MPa is approx. 50% higher than that of ROS (25.4 mm) at 450 MPa. The compressive strength of QI at 361 MPa is slightly lower than that of ROS (25.4 mm) at 440 MPa [105]. Day et al. instead of using ROS, compounded CF-PEEK scrap in a twin extruder with additional neat PEEK to produce SCF injection moulding material. With a resulting average fibre length of 200 µm and 25% fibre weight fraction, the injection moulded samples achieved a tensile strength of 240 MPa and thus outperforming a commercial SCF PEEK material (25% weight fraction, 145 µm average fibre length) [106]. Sarasua et al. investigated the tensile properties of injection moulded recycled CF-PEEK with 10% and 30% fibre weight fractions. Both materials are ground, and subsequently, injection moulded for ten cycles, showing a non-linear decrease in average fibre length from 140 µm (10% fibre weight fraction) and 180 µm (30% fibre weight fraction) to 40 µm after 10 cycles. The tensile strength of the 10% SCF series decreases from 130 MPa after the first cycle to 101 MPa after 10 cycles. The tensile strength of the 30% SCF series decreases from 211 MPa after the first

cycle to 138 MPa after 10 cycles. It is concluded that the decrease in tensile properties is caused by fibre degradation [107].

Apart from fibre orientation and FLD in mechanical recycling of CF-PEEK via compression moulding, the moulding parameters temperature, holding time, consolidation pressure, and cooling rate can significantly influence the mechanical properties. These factors will be discussed in the following sections.

3.2.2. Effect of moulding temperature and holding time

Void formation, viscosity and wettability can be influenced by the temperature and holding time during compression moulding of CFRTP parts. Voids can significantly harm composite parts' mechanical performance, weakening flexural strength, tensile strength and interlaminar shear strength (ILSS) [108]. Enhancing the wettability of PEEK is vital to mitigate the void formation. Improved wettability allows the PEEK matrix to infiltrate the carbon fibre more effectively, reducing the occurrence of voids. Generally, at higher temperatures, the viscosity of the PEEK melt decreases which can result in improved wettability [108, 109]. Hu et al. state that in the temperature range of 360–400 °C, the viscosity of PEEK decreases with increasing temperature but increases with longer holding time, as shown in Fig. 6.

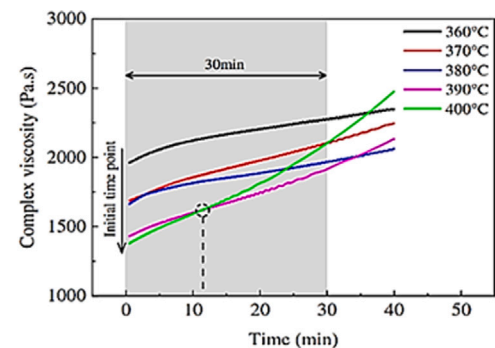


Fig. 6. Viscosity-time curves of PEEK melt under different thermal conditions. Source: Reprinted from [108].

With this behaviour in viscosity, they conclude that wettability also increases with higher temperatures but decreases with longer holding time. The higher the holding temperature, the faster PEEKS' viscosity increases over the holding time. As a result, a similar viscosity of heated PEEK at 400 °C and 390 °C is reached at 12 min [108]. The viscosity increase can be attributed to the long exposure time at high temperatures in which the main chain of PEEK undergoes random molecular chain scission, thermal-oxidative crosslinking, and other effects of thermal degradation, later discussed in Section 3.3. The reduced flow properties due to increased viscosity at long holding times and high moulding temperatures can worsen fibre impregnation. This can result in local resin-rich areas and voids. PEEK has shown the overall lowest viscosity and the best flow properties at a moulding temperature of 390 °C. A longer holding time of 30 min instead of 10 min leads to increased mechanical properties. Moulding temperatures over 400 °C should be avoided to obtain maximised mechanical properties [108].

3.2.3. Effects of consolidation pressure

Besides temperature and holding time, the consolidation pressure affects the void content and the mechanical properties. During CF-PEEK compression moulding, consolidation pressures typically range from 1–10 MPa. The adjustment of consolidation pressure poses a challenge as there is a conflicting objective between achieving a low void content and maximising mechanical properties.

Increased pressure during CF-PEEK compression moulding leads to lower mechanical properties due to decreasing the critical stress and strain at the crack initiation of the composite. This relation is attributed to the accelerated growth rates of PEEK spherulites under pressure, resulting in coarser crystals and a more brittle material [110]. It has

been observed that applying consolidation pressure of > 2 MPa, leads to stagnation and a decline in mechanical properties, particularly in the ILSS [109]. Simultaneously, increasing pressure during the moulding process reduces the porosity of the composite part. A minimum consolidation pressure must be applied to achieve a void content of less than 2%, the highest acceptable void content for high-performance aircraft composite parts. Lystrup et al. demonstrated that by increasing the consolidation pressure from 0.3 to 2.0 MPa, the porosity of the composite part decreases from 13% to 1% [111]. Hu et al. observed a decrease in void content with a successive increase in pressure from 0.5 up to 3.5 MPa [108]. The cited studies utilised recovered or virgin CF fabrics and PEEK films for compression moulding [108,111].

3.2.4. Effects of cooling rate

In addition to the fibre orientation and length, moulding temperature, time and pressure, the mechanical properties of CF-PEEK composites are influenced by the cooling rate. A low cooling rate during moulding leads to a high crystallinity of the PEEK matrix. Furthermore, it results in stronger fibre-matrix interface bonding and thus improved mechanical properties [112]. The positive impact of a slow cooling rate on the crystallinity and mechanical properties is shown in Table 2, which presents the tensile strength values of neat compression moulded PEEK [113].

Table 2

Effect of cooling rate on tensile strength and crystallinity of neat PEEK. Adapted from [113].

Cooling rate [$^{\circ}\text{C}/\text{min}$]	Tensile strength [MPa]	Crystallinity [%]
1	108.5 ± 5.0	38
80	93.8 ± 9.0	30
160	92.5 ± 3.5	28
600	71.7 ± 10.4	26
1000	57.5 ± 3.5	19
2000	53.8 ± 3.3	17

Except for the longitudinal tensile strength and modulus, which are primarily influenced by the properties of fibres, other mechanical properties, such as transverse tensile strength and flexural strength, show sensitivity to the cooling rate. When the cooling rate is increased from 1 to 120 $^{\circ}\text{C}/\text{min}$, the transverse tensile strength of unidirectional CF-PEEK laminate decreases from 100 MPa to 50 MPa, and the flexural strength from 1750 MPa to 800 MPa [113].

Apart from the process parameters, the amount of remelting cycles can impact the mechanical properties. A comparison between mechanically recycled rCF-PEEK with virgin CF-PEEK parts, both compression moulded originally from 20×20 mm virgin strands at the same process parameters, reveals a reduction of 17% in mechanical properties for recycled material [81]. Possible explanations for the decreased mechanical properties, despite comparable FLDs, include diminished CF-PEEK interface bonding and PEEK degradation during remelting. Considering that CF-PEEK parts can undergo multiple life cycles, the effects of repeated PEEK remelting on the mechanical properties and potential adjustment of the process parameters are discussed in the following.

3.3. Effects of compression moulding parameters on CF-PEEK chemistry

PEEK is a thermoplastic that falls under the category of polyaryletherketones. It is a linear homopolymer composed of the monomer shown in Fig. 7 [114].

This etheretherketone monomer is composed of an aromatic main strand that consists of three benzene rings connected by a single bond. Each ring contains three free electron pairs that are highly mobile within the ring. This delocalised electron pair system imparts aromaticity and inertness to the monomer, rendering both the monomer and the polymer chain stable. Since the main strand of the polymer consists only of single bonds, it is not rigid but can rotate around itself at the point between the aromatic groups. This rotation movement occurs

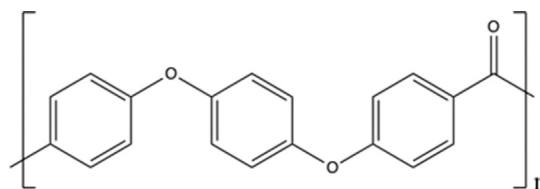


Fig. 7. The chemical structure of polyetheretherketone.
Source: Adapted from [37].

due to thermal energy or external influences such as deformation. This movement becomes significant, especially during the cooling process of the previously molten plastic. As the polymer cools, the chains rotate at the single bonds, forming a crystalline structure. The extent of crystallinity in the resulting material depends on different process variables, which will be discussed in the following sections [114].

When heating CF-PEEK, the PEEK matrix decomposes only when exposed to an oxygen-rich environment above 510 $^{\circ}\text{C}$. Slight residues of PEEK remain in accumulations of fibres or on the fibre surface. The carbon fibres start to decompose at a temperature of 650 $^{\circ}\text{C}$. In a nitrogen environment, the decomposition of PEEK does not begin until 600 $^{\circ}\text{C}$, while carbon fibres do not decompose in a non-oxidative atmosphere at all [115]. During the compression moulding process, changes in the chemical structure of CF-PEEK due to temperatures of up to 400 $^{\circ}\text{C}$ occur. Mechanisms and influences on material properties are reviewed in the following subsections.

3.3.1. Effects on crosslinking density in PEEK

During compression moulding, the chemical structure of the matrix component in CF-PEEK changes depending on process parameters like temperature, time, pressure and atmosphere. Intermolecular reactions occur at keto and ether groups as degradation proceeds. When the material is processed above the glass transition temperature of 143 $^{\circ}\text{C}$, radicals form on the adjacent carbon atom to each ether group [116]. Due to radicals' high reactivity, they tend to form an intermolecular bond with other radicals on the polymer chain. A schematic for the intermolecular mechanism for the formation of crosslinks is shown in Fig. 8 [116]. These crosslinks influence melting temperature, glass transition temperature and crystallinity properties. The number of crosslinks increases with increasing process temperature and time. At constant process temperature, the rate of crosslink formation remains constant. With increasing temperature, more energy is available for forming radicals, thus increasing the rate of intermolecular bond formation [116].

The increasing density of crosslinks causes a decrease in molecular mobility and thus leads to a steady increase in the glass transition temperature [117,118]. The crystallinity behaviour is also affected. Crystalline structures form during the cooling process. Hence, the cooling rate is primarily responsible for the resulting degree of crystallinity. The crystalline content in PEEK decreases, regardless of the cooling rate, if exposed to higher or longer thermal stress before cooling. This is due to the increasing crosslink density, which prevents the material from forming uniform parallel chain arrangements. More crosslinks lower the maximum degree of crystallinity achieved in the material. The reduced maximum crystallinity influences the melting temperature's lowering, as seen in Fig. 10 [119]. Phillips et al. characterised thermal stability of CF-PEEK in air by dynamic rheological analysis, thermogravimetric analysis (TGA) and DSC [118]. They found a clear dependence of crosslinking in PEEK on temperature and time. Furthermore, it is concluded that crosslinking strongly affects the processability of CF-PEEK due to changes in viscosity [118]. Building on the results of Phillips et al. Almeida et al. analysed the changes in viscosity of PEEK due to thermal degradation with the goal of predicting optimal processing conditions for CF-PEEK [120].

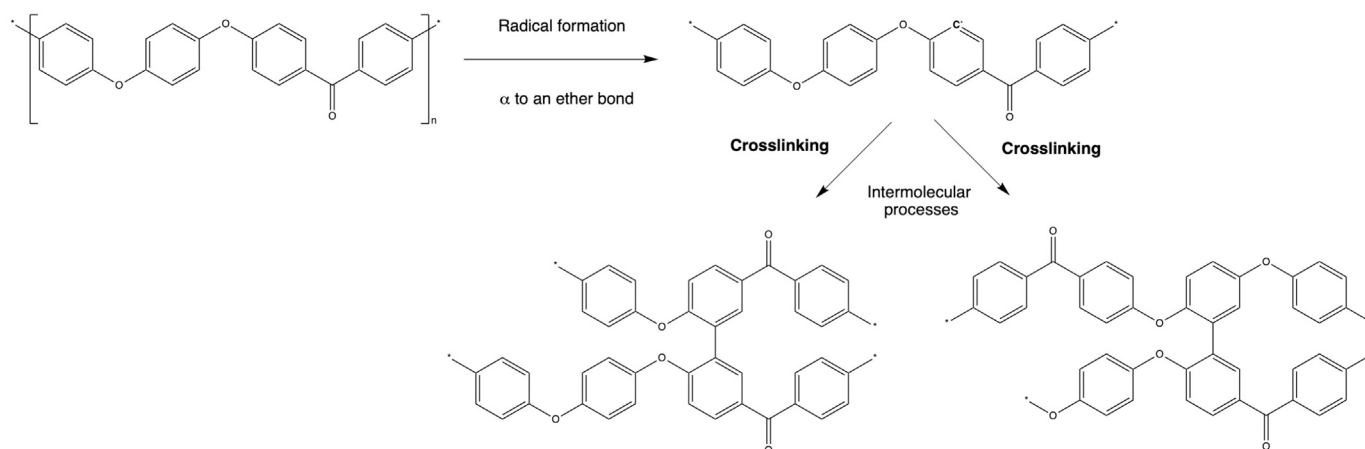


Fig. 8. Intermolecular crosslinking in PEEK during thermal heating via radical formation at the carbon atom.
Source: Adapted from [116].

They conclude that an increase in processing temperature results in increased viscosity due to crosslinking of PEEK. The increased viscosity affects impregnation and eventually leads to higher porosity levels. Whereas longer processing times at 380 °C to 410 °C do not affect consolidation [120].

3.3.2. Effects on ring-opening in PEEK

PEEK is characterised by its thermal stability due to the large number of aromatics in its structure. Aromatic structures are significantly more stable than purely aliphatic polymer chains. Aliphatic structures are chemical compounds that are not aromatic and are composed only of carbon and hydrogen. With the influence of temperature, the number of aromatic compounds in the material continues to decrease. This can be analysed by the Fourier transform infrared spectroscopy (FTIR) based on decreasing intensity of absorption in the benzene region. It can be concluded that ring-opening reactions must take place during thermal treatment in addition to crosslinking reactions [117]. These ring-opening reactions are entropically controlled and thus depend on the degree of disorder in a system. This process starts above the glass transition temperature [121].

New functional groups, such as the hydroxyl group, are formed by the cleavage of the keto group. The number of hydroxyl groups increases with time as ring-opening reactions continue to occur during degradation [117]. The absorbance in the range of carbonyls increases due to the formation of other carbonyl groups, such as esters, aldehydes, carboxylic and peracids, among others. Fig. 9 shows this increase in absorbance in the FTIR analysis results. Mylläri carried out a study whereby PEEK fibre samples with a diameter of $410 \pm 10 \mu\text{m}$ were

aged at a constant temperature of $250 \pm 0.1 \text{ }^\circ\text{C}$ for up to 128 days. The study aimed to characterise the effects of a maximum use temperature of 250 °C on PEEK [117]. Different phenomena, such as ring-opening reactions and the formation of other carbonyl groups, occurring in the material during degradation can be observed [117,122,123].

Due to the increasing loss of the aromatic character, the melting temperature decreases. After a certain point of thermal degradation, the material cannot be molten anymore. The reason for this is that in addition to the melting temperature, the decomposition temperature also decreases. With increasing process time at high temperatures, the difference between melting and decomposition temperatures shrinks from an initial delta around 200 °C to a delta near 0 °C [117].

The ring-opening mechanism itself influences only thermal properties. However, the combination of crosslinking and ring-opening reactions intensifies the change in the material properties. Both decrease thermal stability and lead the material to a thermoset character regarding melting properties. The melting temperature decreases, and the glass transition temperature increases [116]. With increasing process temperature and time, this influence becomes more apparent. A study by Pascual [116] was carried out to evaluate the stability of PEEK in fast processing cycles at high temperatures in the air. PEEK fibres were first thermally stressed in a convection oven at 400, 430, 460 and 490 °C for 30, 60, 180 and 300 s. For the differential scanning calorimetry (DSC), the thermally stressed samples were then run through the temperature program from 0 to 360 °C, 360 to 0 °C and finally from 0 to 400 °C with a constant rate of 10 °C/min. The most significant change in the glass transition temperature can be observed in Fig. 10 at a process temperature of 430 °C with an

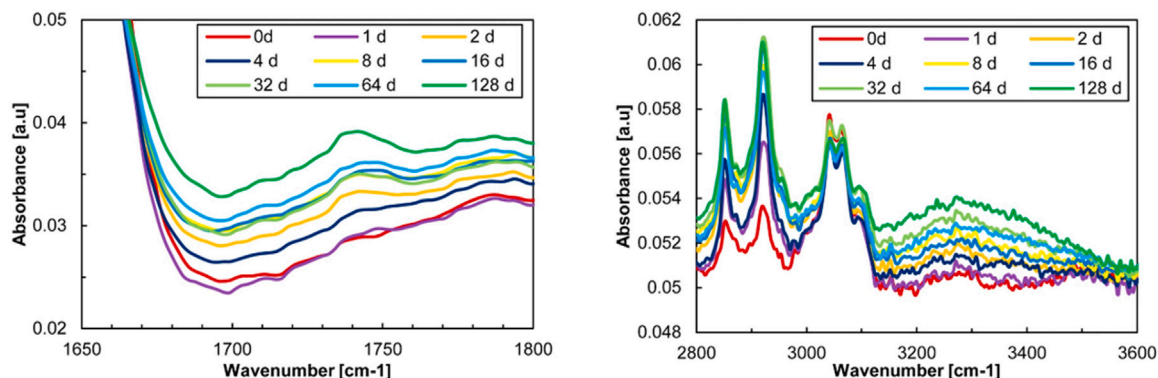


Fig. 9. FTIR spectra of thermally aged PEEK in carbonyl region from 1540 to 1870 cm^{-1} (left) and hydroxyl region from 2800 to 3700 cm^{-1} (right).
Source: Reprinted from [117].

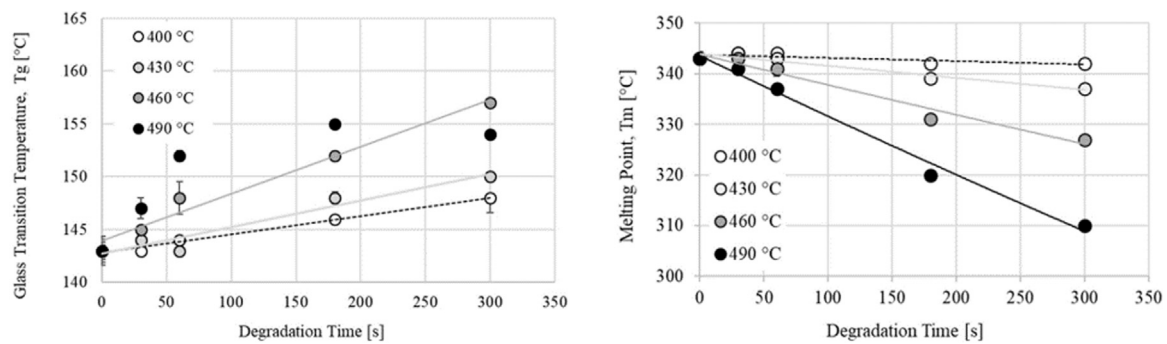


Fig. 10. Effect of process temperature and time on glass transition temperature (left) and melting temperature (right) of PEEK analysed by DSC. Source: Reprinted from [116].

increase of about 15 °C. At 490 °C, a plateau is reached, indicating that short-term stabilisation of the material has occurred through forming a compact protective layer. This type of protective layer is formed due to faster degradation than oxygen diffusion into the material at high temperatures. The material's surface degrades faster than the rest of the sample, forming an oxygen-impermeable protective layer. No further crosslinking or ring-opening reactions occur, and the glass transition temperature plateaus. At low temperatures, on the other hand, the degradation processes proceed slowly, and oxygen has enough time to penetrate the entire material [116].

In addition to the thermal properties, the mechanical properties of the PEEK fibres are affected by longer thermal treatments. The ductility of PEEK decreases rapidly under thermal stress while tensile strength and elastic modulus increase [117]. The increase in the glass transition temperature and Young's modulus can be attributed to the increasing crosslinks [124]. With thermal treatment, except for the increasing brittleness and the decreasing ductility, a short and long-term improvement of the mechanical properties can be observed throughout controlled aging [117]. These results only represent the non-fibre-reinforced PEEK fibres in this study. The influence on the mechanical properties in carbon fibre-reinforced PEEK materials could deviate from these.

3.3.3. Effects on crystallisation characteristics of PEEK

PEEK is a semicrystalline material with a crystalline and an amorphous phase. The crystallinity within the material is decisive for many mechanical and thermal properties and an essential feature of PEEK to be characterised [114].

The cooling rate after heating CFRTP above melting temperatures has a major influence on the matrix structure and the interface. The mechanical and thermal properties of the matrix and interface depend on their crystallinity and, thus, on the cooling rate. Process parameters such as maximum temperature, pressure, and holding time influence the crystallinity less significantly than the cooling rate. At low cooling rates as 1 °C/min, crystalline structures can be formed almost perfectly, resulting in a high degree of crystallinity of 38% [125]. Even a slight increase in the cooling rate to about 7 °C/min induces significant changes in the structure and the mechanical properties [113]. The strength decreases significantly with increasing cooling rate. The tensile strength is affected significantly more in the transverse direction of CF-PEEK than along the fibres. The strength decreases with crystallinity [113]. At cooling rates ranging from 1 to 30 °C/min, which are relevant for industrial compression moulding, the maximum crystallinity remains at 38%–33%. At higher rates of 600 °C/min or 1500 °C/min the crystallinity decreases to 25% or 12%, respectively [125]. The cooling rate can also affect the interphase's structure, where transcrystallisation occurs. Crystals spread on the surface of the fibres in the direction of the heat flow, and the interphase thereby acquires its binding effect between fibres and matrix [126]. In the existing literature, there is a lack of consensus regarding whether transcrystallisation enhances or diminishes material strength [127]. Gao and Kim [113] suggest

that the material's changing structure affects the interphase's bonding strength. In the matrix, with higher cooling rates, the crystalline fraction decreases, which results in a decrease in strength. Above a critical cooling rate of 400 to 600 °C/min, transcrystallisation in the interface is no longer possible and an amorphous interphase forms around the fibre. Thereby, the interlaminar strength decreases significantly. When the critical cooling rate is exceeded, the strength does not decrease significantly further, while crystallinity in the matrix continues to decrease. The lower the cooling rate, the better transcrystallisation is achieved, improving mechanical properties. In contrast, Lustiger [128] concluded that in the presence of transcrystalline interphase, lower interlaminar shear strength and decreased transverse flexural strength were observed. It should be noted that matrix and interphase structures can influence the mechanical properties to the same extent, and accordingly, the influence of both phases must be taken into account. The fibres themselves remain unaffected by the cooling rate [113].

As already described above, crystallinity also is influenced by the crosslinking that occurs during the degradation process. The crystallinity decreases with increasing crosslinking density, which affects not only the mechanical but also the thermal properties of the material. Thermal stability also decreases with decreasing crystallinity, resulting in a decrease in melting temperature and an increase in glass transition and recrystallisation temperature. It should be noted that the influence on thermal stability is more significant in oxygen-containing than in nitrogen-containing environments [119].

3.3.4. Effects on interphase properties

As described in Section 3.3.2, tensile and compressive strength in neat PEEK can improve through tempering at elevated temperatures [117,129]. In Section 3.2.4, it is shown that remoulding can have negative effects on the mechanical properties of CF-PEEK [81]. Assuming that the strength of carbon fibres does not change in the range of 300 °C to 500 °C, this decrease can be attributed to matrix property changes during compression moulding or interphase degradation. The sizing composition mainly drives interphase characteristics. Since the exact composition of sizing material is not given in most studies, only assumptions can be made about the change of interphase with temperature and time. It is known that there is a change in the crystallisation behaviour of PEEK interface, as described above. However, it is unclear whether a transcrystalline interphase has a positive or a negative effect on mechanical properties [127]. Further research has to be conducted in this area.

3.4. Conclusion on the effects of compression moulding parameters on CF-PEEK

Throughout this section, research findings on the effects of mechanical comminution with subsequent compression moulding on CF-PEEK properties have been reviewed. It has been shown that mechanical

Table 3
Effects of compression moulding processing parameters on CF-PEEK.

Parameter	Effect
Fibre length	<ul style="list-style-type: none"> Compressive strength increases with longer fibres [104,105] Tensile strength increases with longer fibres [104,105] Tensile modulus increases with longer fibres [102] Flexural strength increases with longer fibres [104,105] Shear modulus increases with longer fibres [102,103] Shear strength increases with longer fibres [102,103] Waviness decreases with longer fibres [102]
Moulding temperature	<ul style="list-style-type: none"> Temperature influences void formation [108,120] Higher temperatures result in lower viscosity and thus improved wettability or impregnation (in the range of 360 °C–400 °C) [108,109] Higher temperatures result in higher ILSS (in the range of 360 °C–400 °C) [109] Higher temperatures result in lower ILSS (above 420 °C) [109] PEEK begins to decompose above 510 °C in oxygen-rich environment [115] PEEK begins to decompose above 600 °C in nitrogen environment [115] CF begins to decompose above 650 °C in oxygen-rich environment [115] CF does not decompose in non-oxidative environment [115] Intermolecular crosslink density increases with temperature above T_g (143 °C) [116,118,120], resulting in lower T_m [118,119], higher T_g [117,118] and lower crystallinity of PEEK [119] Ring-opening in aromatic PEEK structures above T_g (143 °C) result in a decrease of T_m [116,117,121]
Holding time	<ul style="list-style-type: none"> Longer holding time results in higher viscosity (in the range 360 °C–400 °C) [108,118] Longer holding time results in higher ILSS (in the range of 0–20 min) [109] Intermolecular crosslink density increases with holding time above T_g (143 °C) [116,118] resulting in lower T_m [118,119], higher T_g [117,118] and lower crystallinity [119]
Consolidation pressure	<ul style="list-style-type: none"> Higher pressure results in lower void content [108,111,120] Higher pressure results in accelerated growth rates of PEEK spherulites, resulting in coarser crystals [130] Higher pressure results in lower flexural modulus and flexural strength (in the range of 0–200 MPa) [130] Above 2 MPa, higher pressure results in lower ILSS [109]
Cooling rate	<ul style="list-style-type: none"> Lower cooling rates result in higher crystallinity [112,113,125] Lower cooling rates result in stronger fibre-matrix interface [112] Lower cooling rates (in the range of 1–120 °C/min) result in higher tensile strength [113] Cooling rate has an effect on transcrystallisation on the CF surface with an unclear effect on fibre/matrix interphase properties [113,126–128]

recycling of CF-PEEK with reformatting via injection moulding or compression moulding over several cycles is viable. Mechanical properties can potentially compete with commercial virgin discontinuous fibre-reinforced PEEK. Better mechanical properties are generated with longer fibres. Processing is still possible after several cycles. Besides fibre degradation, there are degradation effects in the PEEK matrix and potentially in the fibre-matrix interface. Fibre shortening has the main effects on mechanical properties and is mainly influenced by the input material or component to be recycled, but also by the settings during comminution and sieving. Chemical degradation effects in the matrix strongly depend on the temperature, atmosphere, pressure and duration of processing. PEEK degradation effects include chain scissoring, ring openings, and crosslinking. Few research findings are available on the effects of CF-PEEK degradation on mechanical properties and optimising processing settings for compression moulding over several cycles. The reviewed research findings on the effects of compression moulding on CF-PEEK properties are summarised in the Table 3.

4. Conclusion

In this paper we reviewed the state of the art in mechanical comminution followed by LFT compression moulding for carbon fibre-reinforced plastics, focusing on CF-PEEK. Several advantages were identified compared to thermal and chemical reclamation routes, particularly the relatively low energy consumption and the preservation of the high-quality thermoplastic matrix, favouring the mechanical comminution. However, fibre shortening is a drawback leading to a loss in the fibre-dominant tensile and compressive mechanical properties compared to continuous fibre-reinforced composites. On the other hand, the relatively high matrix-dominant shear properties resulting from preserving the PEEK matrix could open up new application areas. To enable industrial implementation, it is necessary to predict the mechanical properties of recycled CF-PEEK. A significant decrease is

expected after the first recycling cycle, mainly due to fibre shortening. However, research indicates that further degradation of material properties occurs after subsequent recycling cycles. This degradation results from further fibre shortening and matrix degradation during the shredding and compression moulding. Various degradation mechanisms can be observed in compression moulding, including the formation of crosslinks and ring-opening reactions, which affect the thermal and mechanical properties of the thermoplastic matrix, making remelting and processing more challenging. However, the material properties of compression moulded mechanically comminuted CF-PEEK are not comprehensively quantified. Research must address these research gaps to implement CF-PEEK mechanical recycling route via compression moulding successfully. These gaps include further development of the process chain to increase the technology readiness level, comprehensive analysis of resulting thermochemical and mechanical characteristics and consideration of degradation effects. The optimal process condition must be found to maximise the number of recycling cycles. Bridging these research gaps will be instrumental in advancing the recycling of CF-PEEK in specific and CFRP in general and facilitating its industrial application.

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CRediT authorship contribution statement

Jan Teltschik: Writing – review & editing, Writing – original draft, Visualization, Investigation, Data curation, Conceptualization. **Julius Matter:** Writing – original draft, Investigation, Data curation. **Sven Woebeking:** Writing – original draft, Investigation, Data curation. **Kristina Jahn:** Writing – original draft, Investigation, Data curation. **Yara Borja Adasme:** Writing – review & editing, Validation. **Wim Van**

Paeppegem: Writing – review & editing, Validation. **Klaus Drechsler:** Writing – review & editing, Validation. **Marwa Tallawi:** Writing – review & editing, Validation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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