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REVIEW OF RECYCLING AND TRACEABILITY METHODS FOR CARBON FIBRES

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GLOSSARY/ABBREVIATIONS

CFRP	Carbon fibre-reinforced plastic
CF	Carbon fibre
rCF	Recycled carbon fibre
vCF	Virgin carbon fibre
EU	European Union
H_2	Hydrogen
CH_4	Methane
BHET	Bis(2-Hydroxyethyl) terephthalate
KOH	Potassium hydroxide
CsOH	Caesium hydroxide
NaCl	Sodium chloride
TRL	Technology readiness level
ASTM	American Society for Testing and Materials
ISO	International Organisation for Standardisation
GPa	Gigapascal
mm	Millimetre
K_2CO_3	Potassium carbonate
scPrOH	Super-critical propanol
H_2O_2	Hydrogen peroxide
K_3PO_4	Tripotassium phosphate
mA	Milliampere
E1	Longitudinal Young's modulus
E2	Transverse Young's modulus
ERDE	Explosives Research and Development Establishment
HiPerDiF	High-performance discontinuous fibre
NaOH	Sodium hydroxide
HNO_3	Nitric acid
TEPA	Tetraethylenepentamine
IFSS	Interfacial shear strength
ILSS	Interlaminar shear strength
CVD	Chemical vapour deposition
CNT	Carbon nanotube
LCA	Life-cycle assessment
LCIA	Life-cycle impact assessment
vf	Volume fraction
MJ	Megajoule
Kg	Kilogram
HS	High strength
HM	High modulus
PAN	Polyacrylonitrile
SMC	Sheet moulding compound
BMC	Bulk moulding compound
FDM	Fused deposition moulding
SLS	Selective laser sintering
SEARRCH	Sustainability engineering assessment research for recycling composites with high value
RFID	Radio frequency identification
GPa	Gigapascal

EXECUTIVE SUMMARY

The drive to manufacture lightweight structures has contributed to the use of carbon fibre-reinforced plastic (CFRP) composites in multiple sectors. Unfortunately, this rapid development also creates a problem with waste management as these structures approach their end of life. This creates a necessity to develop recycling methods for carbon fibre recovery and reformatting. This report provides an overview of different recycling methods available for recovering carbon fibres from CFRP materials, as well as an overview of existing methods for establishing the traceability of recycled fibres. Fibre recovery methods are based on thermal or chemical techniques to decompose the matrix from CFRP to liberate the fibres. These recycling methods can have a significant impact on the mechanical properties of the recovered fibre. Thus, testing and validating the quality of the recovered fibres become important processes to undertake to assess the suitability of fibres for reformatting. Test methods such as single fibre tensile tests have been widely used in the existing literature to quantify the tensile properties of recovered fibres. From this study, it has been identified that there are no specific test standards for evaluating the quality of the recycled fibres. As the properties of recycled fibres are heavily influenced by the reformatting techniques, this report aims to highlight the need for suitable reformatting techniques to gain the full potential of recycled fibres. This report provides an overview of existing fibre recovery and reformatting/remanufacturing methods for recycling carbon fibre to establish a circular economy. From review of the literature, it has been identified that existing traceability methods are very limited and require further development. The implementation of material tracking systems and standards to characterise recycled fibres will help to improve the quality of recycled carbon fibres thus encouraging the use of recycled carbon fibres instead of virgin carbon fibres.

1 INTRODUCTION

The demand for carbon fibre-reinforced plastics (CFRP) has increased rapidly over the years and is expected to continue in the future. The aerospace sector remains the number one consumer of CFRP because of its light weighting ability, which has a direct influence on reducing fuel consumption and emissions. For instance, in an Airbus A350 XWB, CFRP constitutes 53% of the materials used in the aircraft. This is a drastic increase when compared to its predecessor (Airbus A380) which had only 23% of composites by weight. Similarly, the demand for CFRP has increased in the automotive sector with electric vehicles targeting light weighting to increase their range. Furthermore, composites are also used in wind turbine blades, hydrogen storage tanks, oil and gas industries as well as in sports equipment. It is anticipated that the demand for CFRP will continue to grow at the rate of 6% per year as per Figure 1 [1].

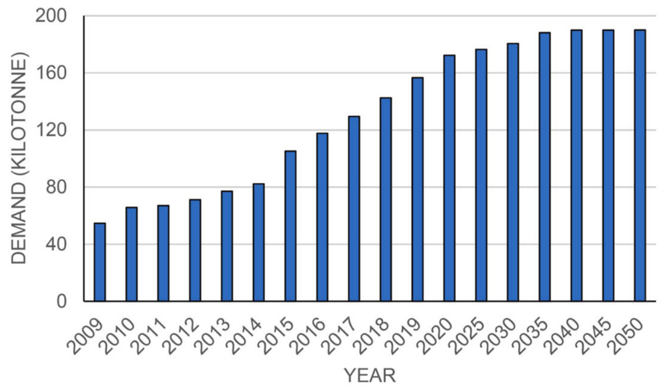

Figure 1: Global demand for carbon fibre [1]

Considering the end of life of aircraft, it is expected that composite scrap from retired aircraft alone will reach 23 kilotons per year and a cumulative waste of 438 kilotons from end of life aircraft and wind turbines by 2050 [2]. This leads to a major challenge in forthcoming years. At present most composite waste is incinerated or goes to landfill. To promote sustainability and to protect the environment, legislation has been imposed by the European Union for landfill (1999/31/EC landfill directive) and incineration (2000/76/EC waste incineration directive). The 2008 EU waste framework directive (2008/98/EC) provides waste management principles promoting reuse and recycling. The recent end of life vehicle directive (EU 2000/53/EC) implements a strict mandate that vehicles disposed of after 2015 should be 85% recyclable. Figure 2 demonstrates the EC waste framework directive which reiterates the order of handling and disposing of waste.

Figure 2: EU waste framework directive [3]

The UK government's composite strategy developed on behalf of the then Department for Business Innovation and Skills identified sustainability and recycling as one of the major goals for the composites industry to ensure that it is environmentally sustainable [4]. This creates an increasing demand to recycle carbon fibre at a commercially larger scale. Lack of market opportunity for recycled carbon fibre, higher recycling costs and inferior mechanical properties of recycled fibres compared to virgin fibres remain challenges for recycled composites [5]. This report provides a brief overview of recycling techniques under development and the traceability of recycled carbon fibres.

2 CARBON FIBRE RECYCLING TECHNIQUES

CFRP materials are made from polymer matrix combined with carbon fibre reinforcement. The strength, stiffness and chemical stability of CFRP makes it hard to recycle [6]. Carbon fibre recycling depends on the type of polymer used as the matrix material. Polymer matrices can be widely classified into two groups: thermosets and thermoplastics [7]. A thermoplastic material can be easily recycled by melting and remanufacturing into new products. However, composites that are manufactured with thermoset polymers, such as epoxy or phenolic resins, become a challenge to recycle because of the 3D cross linked structure [8] that forms on curing of the polymer. Several recycling methods have been studied over the years to recycle thermoset composites; these are summarised in Figure 3. Generally, CFRP recycling involves two processes. In the first process the composite scrap is broken down into smaller pieces using mechanical methods to reduce the size, which are then subjected to thermal or chemical recycling methods for fibre extraction. During the second process, recovered fibres are repurposed in nonwoven mats or other formats of raw material which are then used to manufacture composite components [9].

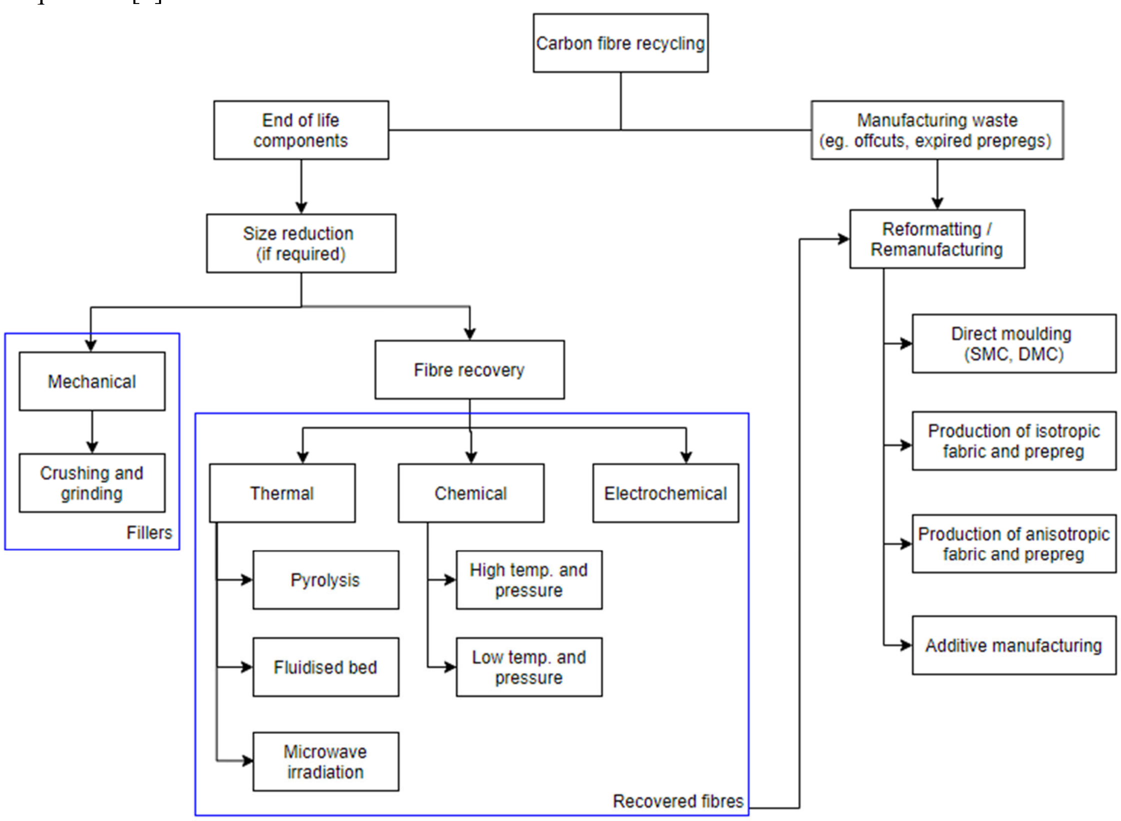

Figure 3 Flowchart outlining different carbon fire recycling techniques

2.1 MECHANICAL RECYCLING

Mechanical recycling is one of the oldest and most commonly used recycling methods for recycling glass and carbon fibre-reinforced plastics. In this technique, the size of the CFRP scrap is mechanically reduced by crushing, milling, and/or grinding. The scrapped pieces are then passed through a sieve to segregate the recyclate depending upon the size of particles. The finest particles after sieving are mostly resin and filler material and the coarser products are more fibrous in nature [10], [11]. The typical size of mechanical recyclates varies from ~100 µm to 10 mm. Figure 4 shows the Wittmann granulator with a processing capacity of 30 Kg/hr and with a constant drum speed of 200 rpm which is commonly used for mechanical recycling of CFRP [12]. Mechanically recycled composites are widely used as fillers along with virgin composite materials, sheet moulding compound (SMC) and bulk moulding compounds (BMC) [10]. Mechanically recycled composites are also incorporated with concrete and asphalt in the construction industry [13].

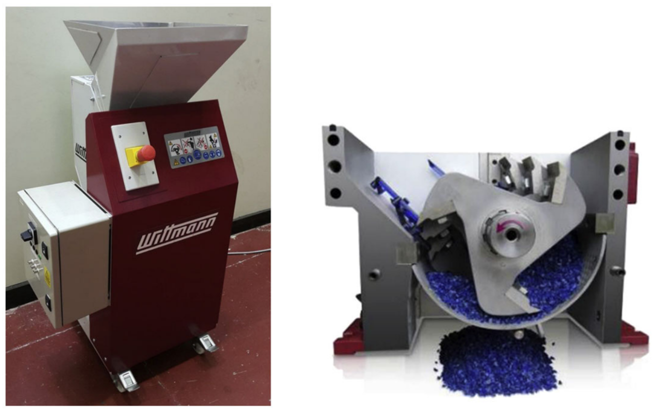

Figure 4: MAS1 Wittmann granulator and rotary cutting chamber used for mechanical recycling [12]

2.2 THERMAL RECYCLING

Thermal recycling is the most widely researched and commercially used recycling technique for recycling CFRP. Unlike mechanical recycling, carbon fibres can be reclaimed using thermal recycling. During a thermal recycling process the polymeric matrix of the CFRP is thermally degraded leaving the reinforcement (recovered carbon fibres) along with other by-products from degraded polymers in the form of liquids and gases. The operating temperature range of thermal recycling processes depend on the matrix material and can vary from 400°C to 750°C. Thermal recycling can be classified into pyrolysis, fluidised bed, and gasification techniques.

2.2.1 Pyrolysis

Pyrolysis involves the matrix material of the composite being thermally degraded at elevated temperature in the absence of air. The process is carried out at elevated temperatures ranging between 400°C to 700°C depending on the matrix material. The thermal resistance of carbon fibres makes the pyrolysis process suitable for reclaiming fibres. Along with reclaimed fibres, this process generates other by-products such as liquid and gases. The liquid by-products which are hydrocarbon in most cases are collected separately and used as a precursor for manufacturing other chemicals. The gases generated, such as H_2 and CH_4 are fed back to the reactor as fuel for combustion [14].

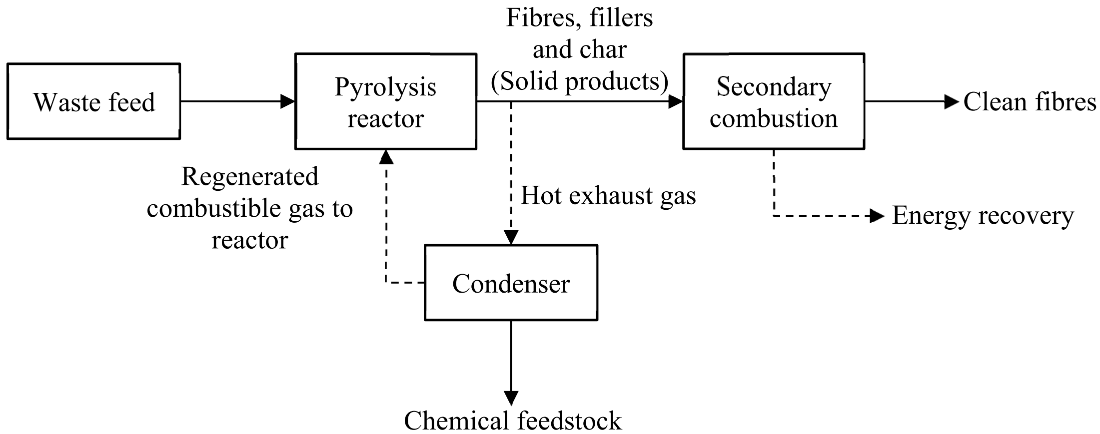

Figure 5: Pyrolysis process for reclaiming carbon fibres from CFRP [15]

Although pyrolysis is one of the most efficient processes for recovering carbon fibres, the recovered fibres are discontinuous, shorter, and fluffier than virgin fibres [15]. When compared to virgin carbon fibre, recovered fibres are known to be of poorer quality [16]–[18]. To improve the quality of the recyclates, pyrolysis combined with different atmospheres has been investigated. The most common methods include pyrolysis in a vacuum, under nitrogen atmosphere and using super-heated steam.

In vacuum pyrolysis, the scrap composite is heated in a vacuum chamber to a temperature of 500°C. This breaks down the resin in the composite liberating the fibres. Then the liberated fibres are processed at a higher temperature of 525°C which removes any residual resin or contaminants in the fibres [19]. Pyrolysis under vacuum is one of the simplest methods of fibre recovery with huge potential for recycling at a commercial scale.

Although pyrolysis under vacuum is one of the easiest methods of recovering carbon fibre from scrap composites, the recovered fibres can contain char or residual matrix material from the process. To prevent char formation and to recover fibres with acceptable surface quality, the process must be carried out in the presence of oxygen. Unfortunately, this can have a detrimental effect on the mechanical properties of the fibre. To prevent fibre damage and to attain a good surface finish, pyrolysis under a nitrogen atmosphere was developed. Yang *et al.* [20] investigated the mechanical properties of recovered carbon fibres under an oxygen-nitrogen atmosphere. It was observed that with 5% oxygen concentration there was a minimal drop in the tensile strength, but when the concentration was increased to 10% the tensile strength of the fibres was significantly reduced.

Pyrolysis under superheated steam is an advanced process which uses superheated steam to decompose the resin from the fibres. Superheated steam is achieved by heating saturated steam over steady pressure. Kim *et al.* [21] investigated the process of recovering carbon fibres using superheated stream. Superheated steam is used to heat the CFRP waste to a temperature of up to 550°C. The steam heats the CFRP recyclates uniformly. Unlike other methods the resin is not vaporised but broken down to tar. This can be collected separately and used as a precursor for manufacturing chemicals. Also, the recovered fibres were reported to have a better surface quality eliminating the requirement for subsequent surface treatment.

Although pyrolysis is one of the most well-developed and commercially advanced processes for fibre recovery, the fibres exhibit a significant loss in their tensile strength when compared to their virgin form [16]–[18]. To a large extent this is due to high temperature and oxidation of fibres during the process.

2.2.2 Fluidised bed

Fluidised bed is a thermal process used to recycle composite materials. This process was developed by the University of Nottingham and can be used to recover glass or carbon fibre [22]. In a fluidised bed process the scrap composites are chopped to an average size of 25 mm [10]. Then the scrap is fed into a chamber with a fine bed of silica (with a particulate size of 0.85 mm) which is fluidised by hot air. The silica bed volatilises the shredded composites, thus degrading the matrix. The whole process happens at 450 to 550°C. Further degradation of the matrix occurs as a result of temperature liberating the fibres. Further combustion of organic materials happens in the secondary chamber at 1,000°C which removes any residual matrix and fillers from the composite. The heat from the secondary chamber can also be used to generate energy. The fibres are separated from the silica using a stainless-steel mesh [22]–[24]. The recovered fibres from the fluidised bed process are short and fluffy, typical of the form widely used in sheet moulding and bulk moulding compounds.

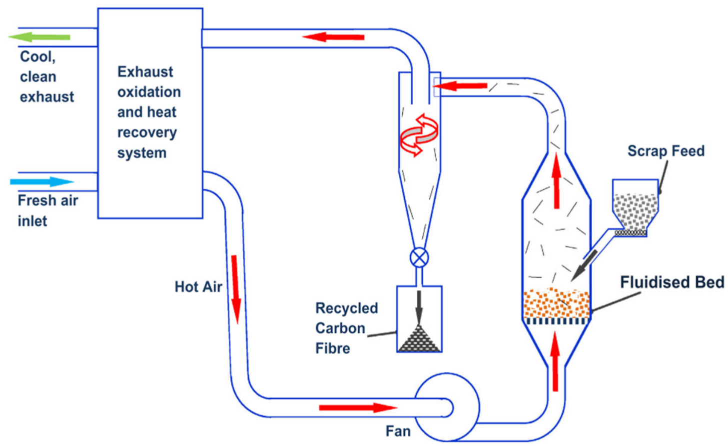

Figure 6: Schematic representation of the fluidised bed process [24]

2.2.3 Gasification

Gasification is a thermolysis process similar to pyrolysis. In gasification the scrap composites are fed into an oxygen free rotating furnace which is maintained at a temperature in the range of 500-600°C [5], [25]. The matrix and any fillers from the scrap composite degrade at the high temperature leaving the reinforcement. As for pyrolysis, the gases and other combustible by-product materials are used as a source of energy for the furnace. After thermolysis, a restricted supply of air is injected into the furnace oxidising the fibres and removing any layer of char from resin breakdown. It is important to control the flow of air to prevent excess damage to the fibres [26]. Gasification can recover cleaner fibres compared to pyrolysis because of oxidation [27] but can have negative effects on the quality of the recovered fibres in terms of mechanical properties. However, the process can be optimised by adjusting the process temperature, time and the amount of oxygen [20], [26], [28].

2.2.4 Microwave irradiation

Carbon fibre recovery with microwave irradiation is a thermal recycling process where conventional heating in a furnace is replaced with microwave heating. Carbon fibre recycling using microwave heating was developed by Lester *et al.* at the University of Nottingham. Lester found that using microwaves for heating is more efficient because the heat originates from the core of the material which leads to faster thermal transfer with minimum energy consumption [29]. It also produces long and clean fibres compared to other methods.

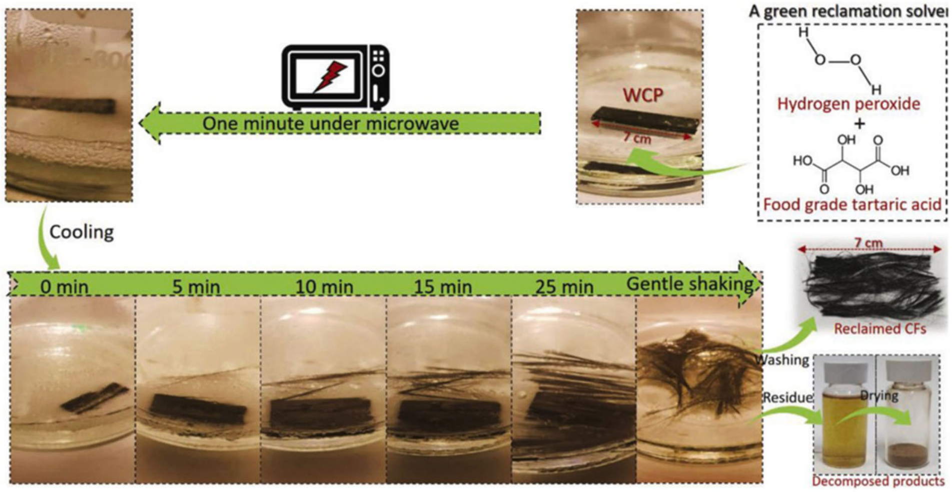

Figure 7: Carbon fibre recovery with microwave irradiation [30]

In a recent publication, Ren *et al.* studied recycling of CFRP using microwave assisted pyrolysis and oxidation giving a yield of 96% recovery of clean carbon fibres [31]. Omid *et al.* used microwaves to recover carbon fibres from scrap composites with a mixture of hydrogen peroxide and tartaric acid as a recycling solvent (Figure 7). It was found that hydrogen peroxide and tartaric acid can initiate epoxy decomposition under microwave irradiation leaving 95% of clean carbon fibres [30]. Although carbon fibre recovery using microwave heating has proved to be efficient, studies concerning the quality and strength of the recovered fibres are very limited.

2.3 CHEMICAL RECYCLING

In chemical recycling, solvents are used to degrade the polymer matrix from the composite scrap liberating the fibres. The polymers from the matrix are broken down to monomers dissolved in the solvent, which provides an opportunity to recycle the chemicals used in the matrix which are then used as chemical feedstock to manufacture polymeric compounds. Due to the nature of recycling by dissolving the matrix material, the process is also called solvolysis. The process can be further classified depending on the solvent used for processing such as hydrolysis, glycolysis, methanolysis, acid digestion and other methods. The process can also be classified depending on the temperature and pressure as sub-critical or super-critical processes [2], [5].

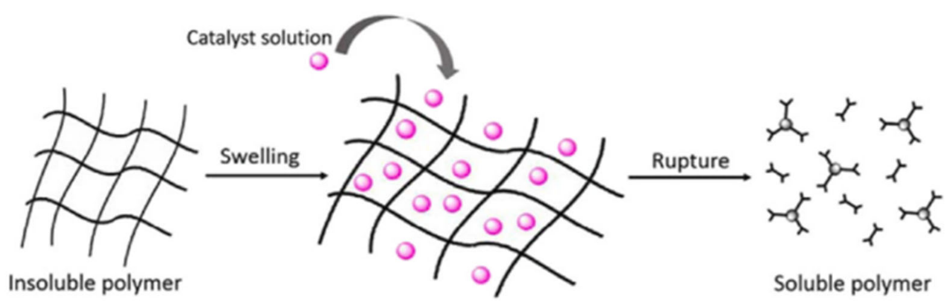

Figure 8: Schematic representation of chemical recycling of CFRP [32]

In glycolysis, carbon fibres are recovered with a solvent with a glycol containing group, like ethyl glycol. Glycolysis can be used to recover fibres from both thermoset and thermoplastic composites. In glycolysis, ethyl glycol reacts with the resin initiating decomposition formulating bis(2-hydroxyethyl) terephthalate (BHET). Upon processing, BHET recrystalises into compounds of isocyanates and maleic anhydrite which can be used to manufacture polyester or polyurethane resins [33]. In a study conducted by Yildirir *et al.,* ethyl glycol was used at 400°C at sub-critical conditions successfully removing 92.1% of the resin from the composite and liberating the carbon fibres [34]. While glycolysis is effective in recovering fibres from scrap composites it can also recover monomers which in turn can be used to recover resins.

Nitric acid is one of the most common solvents used to recover fibre from CFRPs. Liu *et al.* used a nitric acid solution at 90°C to recover fibres from a carbon fibre-reinforced epoxy composite. Nitric acid breaks down the matrix to monomers while the carbon fibres are left undamaged with a tensile strength retention of 98.9 % [35] . Similarly, Ma *et al.* used nitric acid to recover carbon fibres from composites containing an amine cured epoxy resin with a resin decomposition of 99.2%. This proved nitric acid has the potential to recover carbon fibres even from amine cured tough resins [36]. Nitric acid digestion is a well-established method for determining constituent (fibre, matrix, void) volume fractions of composites, methods for which are prescribed in ISO 14127:2008 [37] and ASTM 3171-15 [38]. It should be noted that these standards were not formulated for the application of composite recycling.

Hydrolysis is an another commonly used method to recover carbon fibres from composites. Hydrolysis uses solvents with a hydroxyl group at higher temperature and pressure with the presence of a catalyst. Water and alcohol are commonly used as solvents in hydrolysis. Piñero-Hernanz *et al.* investigated the efficacy of methanol, ethanol, propanol, and acetone as solvents in sub-critical and super-critical conditions to extract fibres from CFRPs at elevated temperatures using sodium hydroxide (NaOH), potassium hydroxide (KOH), and caesium hydroxide (CsOH) as catalysts. The recovered fibres retained 85-99% of the virgin fibre tensile strength [39]. In a recent study Liu *et al.* used water at sub-critical conditions with phenol and KOH to breakdown the epoxy matrix from CFRPs to monophenolic, bisphenolic and amine compounds thus liberating the carbon fibres [40]. Compared to other chemical recycling methods, hydrolysis has the potential to be used on a larger commercial scale as it uses environmentally friendly solvents such as water and alcohol.

From the literature it is evident that chemical recycling methods have the potential to be effective alternatives to thermal recycling. However, it is important to consider that the use of hazardous chemicals such as acids or alkaline substances in large quantities can have a negative effect on the environment. Also, handling large quantities of chemicals can have significant Health & Safety implications. From the available data in the literature [41], [42], the reduction in tensile strength of recovered carbon fibres using chemical method appears to be low when compared to the change in mechanical properties of fibres recovered using other recycling methos.

2.4 ELECTROCHEMICAL METHOD

The electrochemical method is widely used to recycle or separate metals from waste feedstocks. A simple setup for an electrochemical method involves two electrodes immersed in a container with a solution of electrolyte. Sun *et al*. investigated the feasibility of recovering carbon fibre from scrap composite using the electrochemical method. The scrap composite was connected to the positive terminal of a DC power supply and a steel plate was connected to the negative terminal making the anode and cathode, respectively. The anode and cathode were immersed in a vessel containing 3% NaCl solution as an electrolyte [43] – see Figure 9.

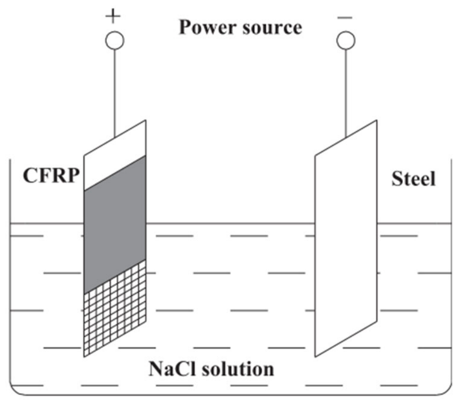

Figure 9: Schematic representation of electrochemical recycling [43]

The electrolysis process was carried out for 21days depolymerising the matrix from the composite. After electrolysis, the carbon fibre obtained was cleaned to remove any residue in an ultrasonic bath with water and alcohol. Zhu *et al.* used a similar electrochemical recycling setup with NaCl as electrolyte and KOH as catalyst to recover 95 % of the carbon fibres from composite waste [44]. The electrochemical method can be more eco-friendly as it generates minimal toxic waste and claims to produce longer and cleaner fibres compared to other methods. The mechanical properties of fibres recovered using electrochemical recycling along with the properties of virgin precursors are discussed extensively in [43].

2.5 ADVANTAGES AND DISADVANTAGES OF EXISTING METHODS

Table 1 provides a summary of the advantages and disadvantages of each recycling method considered in this report along with the associated technology readiness level (TRL).

Table 1: Advantages and disadvantages of different carbon fibre recycling methods

Process	Advantages	Disadvantages	TRL
Mechanical (Section 2.1)	• Simple process • Reclamation of both matrix (cured) and reinforcement • No toxic waste generation.	• Poor quality of recovered fibres. • Loss of mechanical properties. • Irregular mixture of fibres, cured matrix and filler materials makes it harder for remanufacturing.	8-9
Pyrolysis (Section 2.2.1)	• Recovery of long fibres • No use of hazardous chemicals. • Ability to recover chemical feedstock from matrix • Used in current commercial operations.	• Possible formation of char over recovered fibres • Generation of hazardous gas • Mechanical properties of recovered fibres can be influenced by process temperature and conditions.	8
Fluidised bed process (Section 2.2.2)	• No formation of char on recovered fibres. • The process is well documented. • Better suited for contaminated or mixed materials. [9]	• Significant reduction of mechanical properties of recovered fibres. • Matrix material cannot be recovered. • The recovered fibres are short, fluffy, and discontinuous.	4

Gasification (Section 2.2.3)	• Better quality fibres with no char formation. • Recovery of long fibres.	• Process temperature, time and amount of oxygen have significant effect on the quality of the recovered fibres.	3
Chemical (Section 2.3)	• Better retention of fibre properties • Recovery of longer fibres. • Possibility to recover chemicals from matrix material.	• Use of hazardous solvents can impact the environment. • Low tolerance of contaminated precursors. • Not suitable on an industrial scale.	4
Electrochemical (Section 2.4)	• Ability to produce long and clean fibres. • Generates less toxic waste.	• Not economical on a large scale because of the need for large amount of electricity.	3
Microwave irradiation (Section 2.2.4)	• Ability to recover long fibres. • Less damage to recovered fibres. • Energy efficient	• Requires additional cleaning of fibres.	3

3 MECHANICAL PROPERTIES OF RECYCLED CARBON FIBRE

From the literature review it was found that there are no current test standards in place to quantify the mechanical properties of recycled carbon fibres. In most of the existing studies single fibre tensile tests following ASTM D3379-75 [45] or ISO 11566:1996 [46] have been used to compare the properties of recovered and virgin fibres. Although, single fibre tensile tests conducted using existing standards have the potential to estimate the mechanical properties of recycled fibres, the tests can be difficult to perform as the fibre lengths are very short and vary in length. As per ASTM D3379-75, to test the tensile strength of a single carbon fibre, the gauge length of the sample is required to be 2,000 times the diameter. For 7 μm diameter carbon fibres, the gauge length needs to be at least 14 mm which is not always possible with recycled carbon fibres often being much shorter. Table 2 summarises the mechanical properties of recycled fibres obtained using single fibre tensile tests. It can be observed that the gauge length of the fibres varies from 7 mm to 30 mm. Apart from single fibre tensile tests, test methods such as ASTM D4018-17 have been used to characterise the tensile property of recycled fibres [47], [48]. In other studies, ASTM D3039 [49] has been used characterise the tensile properties of recycled composites [50]–[54]. When using ASTM D3039, it is important to mention that the tensile strength measured is for the composite and not of the fibre. An alternate approach to characterise the tensile strength and Young's modulus of single fibres is by using ASTM C1557 [55]. Although, different test methods have been adapted for characterising recycled fibres, it is challenging to address the efficiency of these test methods for use on recycled carbon fibres. This creates a huge uncertainty in the quality of measured data, and necessitates more work to investigate the repeatability and reproducibility of the data when these methods are adapted for characterising recycled carbon fibres.

Table 2: Single fibre tensile tests on recycled carbon fibres

Fibre type	Reclamation process	Fibre length	Tensile strength of virgin fibres (GPa)	Reduction in tensile strength after recycling process (%)	Additional information	Reference
Grafil 34-700	Fluidised bed		4.09	-25%		[56]
	Microwave Pyrolysis			-20%		
	scPrOH			-5%		
Toho-Tenax HTA	Pyrolysis		3.712	-5%		[57]
TR50S carbon fibre from pyrofil	Pyrolysis	25 mm	3.91	-10%	500 °C	[16]
Hexcel AS4	Pyrolysis	10 mm	4.897	-83%	↓ Decreasing temperature	[17]
				-75%		
				-67%		
				-2%		
		20 mm	4.551	-85%	↓ Decreasing temperature	
				-81%		
				-76%		
				-4%		
Torayca T800S	Pyrolysis	10 mm	5.9	-17%		[18]
				-17%	Post treated in 450 °C	
				-69%	Post treated in 600 °C	
				-24%	Post treated in nitric acid	
		20 mm		-24%		
				-69%	Post treated in 450 °C	
				-82%	Post treated in 600 °C	
				-44%	Post treated in nitric acid	
Diverse fibres recycled together	Pyrolysis	10 mm	-22% when compared to standard aerospace prepreg T700 from Toray		400 °C	[58]
Toray T600S	Fluidised bed	10 mm	4.84	-34%	550 °C	[59]

Toray T700S	Fluidised bed	10 mm	6.24	-54%	550 °C	
Hexcel AS4	Fluidised bed	10 mm	4.48	-38%	550 °C	
Grafil MR60H	Fluidised bed	10 mm	5.32	-51%	550 °C	
HexPly® F593	Thermolysis and gasification		3.53	-28%	500 °C	[60]
Carbon fibre	Solvolysis	-	-	-10%		[61]
Toray T600	Super-critical water		4.09	-11%		[62]_
Grafil 34-700	Sub-critical and super-critical alcohol and acetone		-	-15%		[39]
Carbon fibre	Super-critical water	-	-	-1.80%	With acidic catalyst	[63]
				-4.10%	Without acidic catalyst	
Toray T300	Super-critical water and Oxygen	30 mm	3.11	0%	at 95.5% purity	[64]
				-38%	at 100 % purity	
Carbon fibre	Liquid sub-critical water	-	-	-12%	without catalyst	[65]
		-	-	-17%	With K_2CO_3 as catalyst	
Carbon fibres	Super-critical methanol	-	-	-9%		[66]
Toray T300B	Sub-critical benzyl alcohol + K_3PO_4	25 mm	-	0%		[67]
Carbon fibre	Sub-critical water	20 mm	2.6	2%	315 °C + Phenol + KOH	[41]
IM7	Super-critical water + KOH	25 mm	5.25	7%		[68]
Toray T 300	Super-critical water			0 to 10% loss of tensile strength	Processed at 350 °C	[69]
Toray T 700	Solvolysis (Pre-treatment in acetic acid + acetone / H_2O_2)		2.81	-5%	at 80 °C	[42]
				-7%	at 100 °C	
				-13%	at 120 °C	
Toray T 700	Electrochemical method	7 ± 1 mm	-	-20 %	3% NaCl and 25 mA	[43]
				-37 %	3% NaCl and 20 mA	
				-47 %	3% NaCl and 4 mA	

From Table 2 it is evident that recycled carbon fibres experience a significant reduction in their tensile strength compared to virgin fibres. The reduction in tensile strength can be between 2% to 85% depending on the recycling method and atmosphere used. The data in Table 2 also show that for some chemical recycling methods the tensile strength of the fibres has increased by up to 7%. Such studies see inconsistent and have failed to address the reason behind the increase in the tensile strength of recycled fibres compared to their virgin form.

3.1 FACTORS AFFECTING MECHANICAL PROPERTIES OF RECYCLED FIBRES

From Table 2 it is evident that recycled fibres can suffer a significant reduction in their mechanical properties with the exception of a few studies using the chemical recycling method that show little to no change in tensile strength. The mechanical properties of composites are highly dependent on the strength and stiffness of the fibres, adhesion between the matrix and the fibre, the length of the fibre for effective stress transfer and tailored fibre orientation as per principal stress direction [70]. Therefore for composites made with recycled fibres to have acceptable properties it is essential for the recycled fibres themselves to be of suitable quality. There has been much focus on developing processes to improve the form and mechanical properties of recycled fibres; the following sections describe efforts to improve fibre alignment and surface finish.

3.1.1 Fibre alignment

Fibre alignment is a major factor when it comes to retaining the mechanical properties of recycled carbon fibre composites. Recovered fibres are often short and fluffy. Fibre alignment is not only necessary to maintain high E1/E2 ratio but also to increase fibre packing (volume fraction) [71]. A virgin carbon fibre prepreg usually has a fibre volume fraction of 55-60% whereas using recycled fibres in a bulk moulding process a volume fraction of 10% is typically achieved. A higher volume fraction and alignment can be achieved by compressing the fibres with higher pressure but, this has a substantial effect on the fibre length [72]. To improve the fibre alignment several methods have been developed and are broadly classified as dry and wet alignment.

Dry alignment methods use magnetic or electric fields to align the fibres, whereas wet alignment utilises hydrodynamic methods. Timbrell *et al*. found that magnetic fields can be used to align short carbon fibres in lengths of up to 500 μm when suspended in a liquid [73]. Later Yamashita *et al*. used a magnetic moment to align short carbon fibres coated with nickel in a liquid resin matrix. It was found that fibre alignment became difficult to control when the volume fraction of the fibres was more than 5% [74]. Like magnetic alignment, several attempts have been made to align carbon fibres using an electric field. In recent work by Anil *et al*. the efficacy of aligning fibres using an alternating current electric field has been investigated [75]. It was found that electric current can be used to align very short fibres (sub-millimetre length), however as the fibre length increased it prevented the fibres being aligned. Therefore, dry alignment methods are still under development and have not been successful on a commercial scale.

Wet fibre alignment is based on the theory that a slender object with high aspect ratio tends to align itself to the direction of the flow when the fluid flow is laminar. In the case of fibre alignment, the fibres tend to align preferentially towards their longitudinal axis in the direction of the flow [76]. Flow induced fibre alignment has been investigated by various authors for short fibres out of which the process developed by Explosives Research and Development Establishment (EDRE) UK, has produced high alignment of fibres with fibre volume packing up to 50% (fibre packing density). The process follows several stages which include extrusion, filtration, and centrifugal processes. In the EDRE process the fibres are suspended in a fluid medium which is then passed through a converging nozzle. The fibre suspension is sprayed through the nozzle onto a perforated rotating drum where the fluid medium is removed by centrifugal force [72].

The recent development of the High-Performance Discontinuous Fibre (HiPerDiF) technology by University of Bristol has enhanced the capability to produce highly aligned fibre composites. In the HiPerDiF process, the fibres are suspended in a low concentration of water. The suspended fibres are then driven with a pump through a nozzle that directs the fibres towards the orientation plate. When the fibres hit the orientation plate the direction of the fibres changes from transverse to the waterjet direction. When the fibres fall on the perforated conveyer belt, the water is sucked through the pores and the fibres align to the direction of the conveyer [51]. The HiPerDiF process has been successful in aligning 67% of the fibres within ± 3° of the intended direction. Composites manufactured using the HiPerDiF process exhibited a volume fraction of 55% [77]. From the literature, it is evident that the wet fibre alignment technique has strong potential to improve the quality of recycled carbon fibres, however most of these techniques are still in the initial development phase and require further development to be attain higher TRL levels.

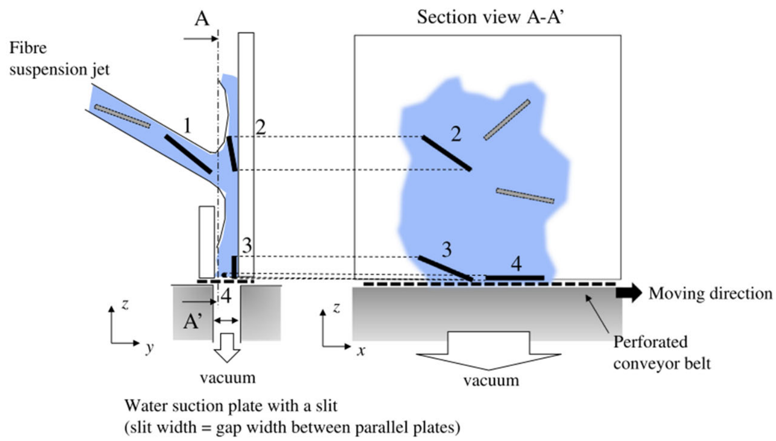

Figure 10: Concept of fibre alignment in HiPerDif [77]

3.1.2 Fibre Surface

The fibre surface quality is an important parameter which directly affects the mechanical properties of composites as it contributes to an effective level of adhesion between the matrix and the fibre. During the recycling process, fibres are exposed to harsh environments such as heat or chemicals depending on the recycling method. This can have a significant effect on the fibre surface quality. The process for improving interfacial adhesion between the matrix and fibres has been investigated for decades. Based on the available literature, the fibre surface modification methods can be widely classified into chemical oxidation, electrochemical oxidation, surface functional group modification by grafting and plasma treatment [78], [79]. Each of these surface modification techniques are briefly discussed in the following section.

Chemical oxidation refers to oxidising the fibre surface using gaseous or liquid reagents. In gaseous oxidation, gases such as oxygen, air, ozone etc are used to oxidise the surface of the fibres. It has been identified that gaseous oxidation is less effective since the process is carried out at high temperatures (350 - 600°C). This increases the chance of fibre pitting, which can have a significant effect on their properties. Oxidation at high temperatures can also cause spontaneous ignition of fibres. However, it has been observed that gaseous oxidation of fibres can increase the fibre surface area and surface rugosity [78] which improves the interlaminar shear strength (ILSS) of the composite. Chemical oxidation treatment using liquid reagents such as acids, NaOH, HNO_3, etc has been investigated using virgin fibres to increase the surface adhesion. In a study conducted by Pittman *et al.*[80] the carbon fibres were immersed in 70% concentrated nitric acid at 115°C followed by reacting with tetraethylenepentamine (TEPA) to generate surface acidic functional groups. The reaction increased the surface area of the fibres which in turn increases the adhesion of fibres to epoxy resin matrices. Similarly, Wu *et al.* [48] studied the effect of surface oxidation of virgin T-300 fibres using aqueous NaOH solution. It was found that treating fibres with NaOH caused higher weight loss of the fibres (loss of surface graphite from fibres) and also increases the surface area of the fibres. This increased the interfacial shear strength (IFSS) and interlaminar shear strength (ILSS). Even though the process increased the IFSS and ILSS of the fibres, the tensile strength of the fibres exhibited a significant decline after the treatment.

Surface oxidation of fibres can be also achieved by using electrochemical methods. In this method the fibres are processed in an electrochemical bath. The electrolytic bath contains caustic chemicals such as inorganic acids, potassium permanganate, sodium dichromate and others. When compared to gaseous oxidation, electrolytic surface oxidation causes minimal pitting and damage to fibres. Also, the electrochemical method is much faster, and the fibre surface property is uniform across all fibres [78]. Donnet *et al.* [81] investigated the effect of electrochemical oxidation on T-300 fibres using NaOH as the electrolyte and nickel as the cathode. After the electrolytic process, the IFSS increased by 67% and the ILSS increased by 10.5%. The properties of treated fibres depend on the current density, oxidation time and the temperature of the electrolyte.

Plasma treatment is one of the proven methods for surface treatment of virgin carbon fibres. During the plasma treatment process, the plasma produces ions, free radicals and meta-stable species which induces ablation, cross linking and oxidation reaction in the fibres [82]. Plasma treatment is one of the widely preferred methods of surface treatment as it does not cause any degradation to the bulk properties of the fibre. Lee *et al.* [79] studied the effect of plasma treatment on carbon fibre recovered from aircraft parts using a chemical method. From the study it was evidenced that plasma oxidised the functional groups on the fibre surface and led to better adhesion between the matrix and reinforcement when compared to untreated fibres.

Surface functional group modification by grafting is another way of modifying surface characteristics of carbon fibre by bonding carbon nanotubes, amines, or polymers on to the surface of the fibre. Multiple surface grafting techniques are reported in the literature for processing virgin carbon fibres out of which surface modification by Chemical Vapour Deposition (CVD) is more common. Qian *et al.* investigated the efficacy of grafting carbon nanotubes (CNTs) to the surface of virgin carbon fibres using CVD. It was found that CNTs increased the IFSS by 26% [83]. Although grafting functional materials has proved to increase the quality of the carbon fibre, implementing surface treatment processes such as grafting may not be financially viable.

4 LIFE CYCLE PERSPECTIVE OF CARBON FIBRE RECYCLING

Life Cycle Assessment (LCA) is a holistic approach used to identify the environmental impacts of a product or process throughout the entire life cycle. The LCA approach takes into account all of the inputs (raw materials, energy, water *etc.*) and outputs (final product, by-products, emissions, waste, *etc.*) from raw materials extraction through to manufacture, distribution, and usage to end-of-life. The outline methodology is defined by the international standards for Environmental Management Systems ISO 14040 and 14044 [84][85].

The LCA methodology has been used widely in the field of composites to investigate and compare the environmental impacts of conventional materials with CFRP or recycled CFRP [86][87][88]. Manufacturing of CF is known to be a very energy intensive process with emission of hazardous by-products [89]. For example, the calculated embodied energy for CF is 183-286 MJ/kg, compared with 13-32 MJ/kg and 110-210 MJ/kg for glass fibre (GF) and stainless steel, respectively [90]. It has been reported, that from an energy perspective, the feasibility of using carbon fibres in automotive applications can only be achieved through higher fuel economy by reducing the gross weight of the vehicle. Using recycled CF is recommended to reduce the embedded energy in automotive parts where fuel economy cannot be achieved [91]. Cradle-to-grave LCA studies of CFRP applications can demonstrate the environmental benefits of recycled CF compared with end-of-life treatment options such as landfilling or incineration [92], [93]. However, comprehensive LCA studies for CFRP applications, where different recycling technologies are considered and address the entire life cycle of CF, are very difficult to find in the literature.

While some studies conclude that using recycled CF in automotive applications is environmentally beneficial compared to using virgin CF [94], some argue that environmental savings depend on the recycling method chosen which may also affect the mechanical properties of the recycled fibres and the weight saving in comparison to virgin CF [95]. Table 3 contains energy consumption data for several recycling technologies that have been developed. Recycling CFRP and using recycled CF has a range of potential advantages, challenges and opportunities and the environmental benefits and impacts resulting from different recycling methods need to be studied further.

Table 3: LCA data associated with recycling of carbon fibre via different methods

Fibre type	Recycling method	Energy requirement (MJ/kg)	Additional information	Reference
Carbon Fibre/Epoxy CFRP – 55% v_f	Pyrolysis	37.36	1 kg of CFRP waste resulted in 0.62 kg of rCF Including transport 0.27 MJ/kg	[96]
	Fluidised Bed	10.25		
	Chemical	38.39		
CFRP	Mechanical	0.27-2.03		[24]
	Pyrolysis	3-30		
	Solvolysis	19.2		
	Chemical	60-90	Study based in Japan compared to 198-595 MJ/kg for vCF production	
CFRP	Chemical	7.62	Study based in South Korea in 2010. Data is from SimaPro	[97]
	Pyrolysis	47.88		
CFRP waste	Microwave Pyrolysis	10		[29], [98]
CFRP waste	Solvolysis	9.2		[99]
CFRP waste - 60 % carbon fibre	Pyrolysis (without heat recovery)	15-30		[100]
	Microwave Pyrolysis	10		
	Thermolysis using steam and nitrogen gas	71.64	Shows environmental advantages compared to waste landfilling according to Life-cycle impact assessment (LCIA) results	

CFRP (Toray T700) with 60% v_f bound with epoxy	Electrochemical	~15	Calculated for CFRP strip (30 mm x 150 mm x 20 mm) with 2000 kg/m^3 density in 3% NaCl solution at a depth of 0.7 mm with a current of 4 mA and voltage of 2.6 V for 21 days	[43]

There is significant variability and discrepancies between LCA data for carbon fibre production in the literature. Scientific papers quote different values for energy consumption in carbon fibre production which range from 200-1,000 MJ/kg with lack of detail in the type of fibre produced (HS, HM, PAN precursor etc) [24], [91], [101], [102]. As presented in Table 3, the energy consumption associated with different recycling methods ranges from 1-100 MJ/kg; when these data are compared qualitatively, recycling processes tend to use less energy than virgin carbon fibre production. Choosing the least energy intensive recycling process to fit the application would be environmentally beneficial over virgin fibre production, not only by saving energy and reducing emissions, but also by reducing the amount of material entering landfills and thus promoting a circular economy.

5 CARBON FIBRE RECOVERY AND REFORMATTING

5.1 RECOVERY FROM DRY WASTE

Waste generated during composite manufacturing constitutes to 40% of the total composite waste [1]. Dry carbon fibre waste products include offcuts, defective material, end of roll fibres, expired prepregs and prepreg trimmings. Traditionally this type of waste has ended up in landfills, however more recently these products have gained the attention for recycling because of their economic value [26]. The advantage of using production waste to recover carbon fibres from is that it can be easily traced back to the virgin fibres and intended properties [103]. Besides the degree of traceability, carbon fibre production waste can easily be spun or combined with virgin fibres which makes it less challenging to recycle when compared to end-of-life components which require special processing. The carbon fibre recovered from production waste can usually be spun into new carbon fibre yarns, fabricating non-woven mats and prepregs. Moreover, in a recent publication by Bruijn *et al.* access door panels from a rotorcraft were replaced (and flown) with a door panel manufactured from recycled carbon fibre from production waste, thus promoting the possibility of using recycled carbon fibre in the aviation industry [104].

5.2 CARBON FIBRE REFORMATTING

The final phase of carbon fibre recycling is remanufacturing new components with recycled fibres. It is important that prior to the remanufacturing stage, the recovered fibres are treated and reformatted into usable forms. The recovered fibres are often fluffy and shorter in length, therefore the manufacturing techniques used for virgin fibres must be modified to suit the recycled fibre. Recycled carbon fibres can be remanufactured as composites or reformatted into textile reinforcements by direct moulding, woven into isotropic or anisotropic textiles or produced by additive manufacturing. Each of the methods is explained in detail in the sections below.

5.2.1 Direct moulding

Direct moulding is a process where the recovered fibres are directly mixed with resin and moulded into a desired part. The common formats when moulding carbon fibres are injection moulding, sheet moulding compound (SMC) and bulk moulding compound (BMC).

In injection moulding the recovered fibres are mixed with the matrix and injected into a preform. Connor investigated the mechanical properties of test coupons manufactured with recycled carbon fibres using injection moulding and compared it with test coupons which were injection moulded with virgin fibres. It was observed that the recycled CFRP exhibited poorer tensile properties but had better impact resistance compared to virgin CFRP coupons [105]. Because of the random orientation of fibres in injection moulded components, the material format is more suitable to the manufacture non-structural parts rather than load bearing structures.

Sheet Moulding Compounds (SMC) are a special form of prepreg where the fibres are mixed with resin and are made it into sheets or rolls. SMC is usually made with chopped glass fibres and polyester resin [106]. These sheets of prepregs are then compression moulded to form the desired part. SMC is becoming increasingly popular in the automotive sector due to its bulk properties and low manufacturing cost. Replacing glass fibres in SMC with recycled carbon fibres will provide distinct advantages such as light weighting and increased stiffness. Palmer *et al*. investigated the mechanical properties of SMC produced with recycled carbon fibres. It was reported that under careful control of volume fraction SMC with better mechanical properties than conventional SMC can be produced. This enables the ability to replace the SMC currently being used in automotive applications with SMC manufactured with recycled carbon fibres [107].

Bulk Moulding Compound (BMC) also known as Dough Moulding Compound (DMC) is a mixture of resin and short glass fibres which have a clay like consistency that can be moulded or extruded into a desired part under compression moulding [106]. Like SMC, BMC has become more popular in the automotive industry for its light weighting capability. Turner *et al* [71] investigated the development of BMC with recycled carbon fibres and compared it with commercial BMC with glass fibres. It was found that BMC made with recycled carbon fibres can be produced at a similar cost of BMC with virgin glass fibres. It was found that BMC with recycled carbon fibres at low volume fractions (less than 10%) has shown similar mechanical properties when compared to BMC made with virgin glass fibres. Also, Saburow *et al.* investigated the mechanical properties of BMC manufactured with recycled carbon fibres and compared it with SMC produced with virgin carbon fibres. It was found that mechanical properties including tension, bending and impact resistance were of the same order of magnitude when compared to data obtained from virgin carbon fibre SMC [108].

5.2.2 Production of isotropic fabrics

To achieve a closed loop application for carbon fibre recycling, it is important to reformat the recovered fibres as reinforcement for CFRP. One established method of reformatting reinforcements is by manufacturing them into non-woven reinforcements. The non-woven reinforcements can exhibit varying levels of anisotropy depending upon the manufacturing process. The reformatting of non-woven fabrics from recycled carbon fibre can be widely categorised in to wet-laid and dry-laid processes.

In a wet-laid process the recovered fibres are suspended in an aqueous medium to align the fibres and to be repurposed as reinforcement. This wet-laid process for carbon fibre recycling is based on a modified papermaking process [109]. The recovered fibres are suspended in a tank with water or aqueous medium to form a slurry which is then pumped in to a headbox which deposits the fibres on to a conveyer belt. The aqueous medium is then drained, and the fibres take the form of a non-woven reinforcement. With the addition of fillers and resin to the process, reinforcements with multiple functionalities can be manufactured. Properties such as areal density can be modified with minimal adjustment to the manufacturing process. The fibre alignment in the reinforcement produced by wet-laid process is random and the material is isotropic [110].

In an air-laid process the fibres are uniformly dispersed by a stream of air on a conveyer belt, where the fibres form a web like structure. This results in formation of isotropic non-woven textile. Unlike the wet-laid manufacturing process the deposited fibres can be fluffy which are then interlocked by a mechanical needle piercing process. Different fibre deposition techniques have been reported in the literature, which can be classified as free fall, using compressed air, closed air circuit and air suction [110]. When compared to the wet-laid process, air-laid can have several limitations including fibre clumps in the airstream which can be difficult to separate, and which can cause defects in the manufactured fabric.

5.2.3 Production of anisotropic fabrics

One of the important features of composite materials is the ability to take advantage of anisotropy to tailor properties required for a particular application. Therefore, it is important to be able to remanufacture composite materials utilising recycled fibres into anisotropic formats. Different remanufacturing methods have been investigated and the most common methods are wet-laid, carding and spinning yarns. Although the wet-laid process is predominantly used for manufacturing isotropic fabrics, it can also be used to produce reinforcements with anisotropic properties. In the process of remanufacturing anisotropic fabrics, the principle of hydrodynamics is used for fibre alignment. This has been discussed in detail under section 3.1.1 of this report.

Carding is a mechanical process which can be used to reformat recovered carbon fibres. In the carding process, the fibre clumps known as bales are broken up to manageable fibre tufts using a bale breaker. The broken fibres are passed through a bale picker to remove any contaminants from the fibre. The fibres are passed through a series of sawtooth rollers known as beaters. Beaters are used to mix the fibres and can also be used to remove any entanglements in the fibres. Resin or any other required additives such as fillers can be added to the fibres at this stage. In the next step, the fibres are rolled over a main cylinder and passed through a series of worker and skipper rollers [110]. The worker and skipper rollers operate in pairs to mix and rearrange the fibres as per the desired orientation. Increasing the number of skipper and worker rollers can improve fibre disentanglement but can have a poor effect on fibre length, which can impair the mechanical properties of the manufactured fabric [111]. The web obtained is passed through the doffer roller. In the doffer zone, the spun web is transferred from the main cylinder which is then removed carefully. Carding is preferred to manufacture fabrics with low areal density because if the fabric becomes thick the sawtooth on the cylinders can become ineffective, leading to defects in the manufactured fabric [112]. The process of carding for reformatting recycled carbon fibre is shown in Figure 11.

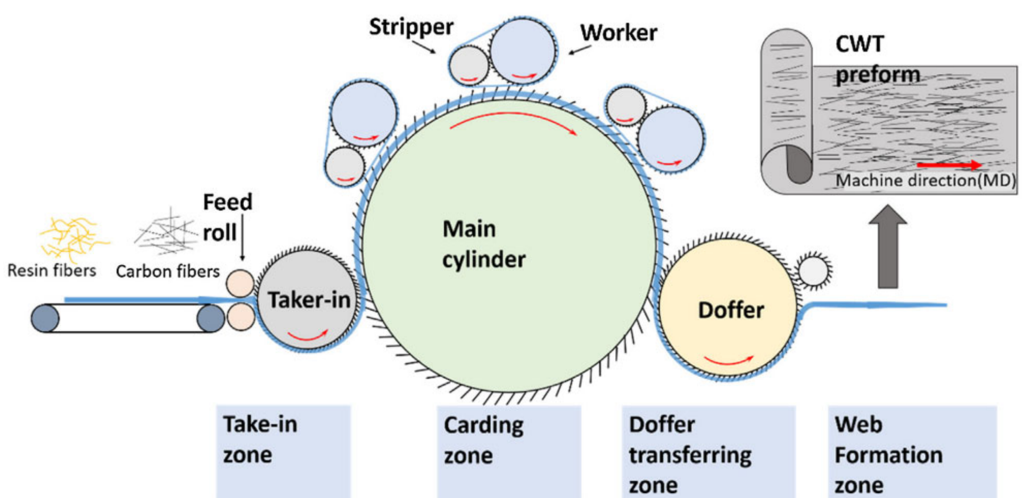

Figure 11: Carding process [112]

Spinning yarns with recycled carbon fibre is another method for manufacturing highly anisotropic carbon fibre fabric. Spinning yarns is often combined with the carding process. During the carding process, the recycled fibres are disentangled and aligned. Then the carded web is transformed to slivers by drafting, drawing and spinning stages to manufacture recycled carbon fibre yarns [90]. In the available literature, dry carbon fibre waste was predominantly used for yarn spinning. Yarn spinning of dry carbon fibre can be more challenging than conventional yarn spinning, because dry carbon fibre waste can exhibit properties like lack of shear resistance, low elongation, and brittle behaviour [113]. This makes it harder to be processed into yarns. Therefore, thermoplastic fibres such as PA6, PP and polyester can be added to carbon fibres to manufacture hybrid yarns. Figure 12 shows the process of manufacturing recycled carbon fibres into yarns. Hengaterman *et al.* have carried out extensive research on manufacturing carbon fibre yarns from recycled carbon fibres [114]–[117]. The initial findings suggest that the fibre length, orientation and mixing ratio with thermoplastic fibres contribute significantly to the properties of the manufactured sliver [114]. Upon further analysis it was found that factors such as yarn twist and volume fraction play a significant part in the tensile properties of the sliver. Also, the recycling process such as pyrolysis can have a detrimental effect on properties such as brittleness and fibre to fibre adhesion (bonding with adjacent fibres) [115]. In another publication, Hengaterman *et al.* [116] investigated the sizing effect of recycled carbon fibres in spinning into hybrid yarns (recycled carbon fibres with polyamide). It has been reported that shortening of the fibres can be caused by the carding process and alternate approaches must be applied to prevent damage to the fibres.

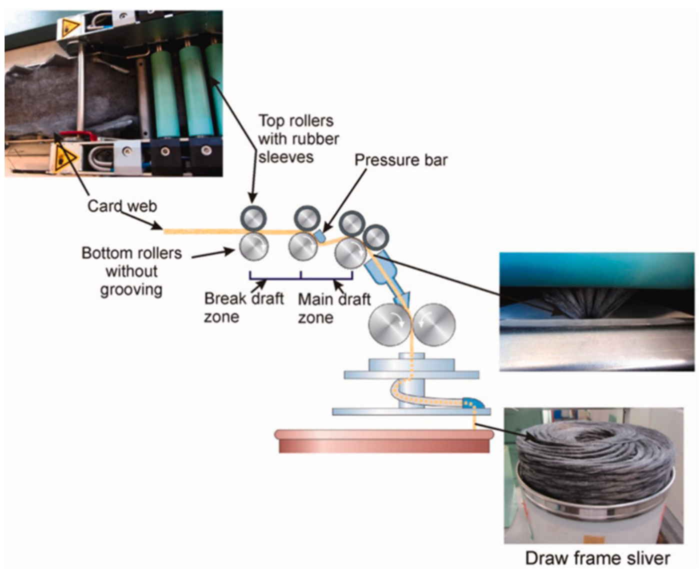

Figure 12: Drawing slivers from recycled carbon fibre [114]

5.2.4 Additive manufacturing

Additive manufacturing, commonly known as 3D printing, is a process of manufacturing where the product is printed in a layer-by-layer form to a desired size and shape. Although, additive manufacturing is known for its ability to manufacture complex shapes and geometries, layer-by-layer printing can result in poor mechanical properties of the final product. Adding carbon fibre to 3D printed parts can improve the mechanical and functional properties [118]. From the published literature it is evidenced that adding carbon fibres to additive manufacturing can improve mechanical properties. Ning *et al.* investigated the mechanical properties of 3D printed plastic specimens with and without carbon fibres. It was reported that the test coupons with carbon fibres exhibited better flexural properties with an 11.82% increase in flexural strength, 16.82% increase in flexural modulus and 21.86% increase in flexural toughness [119]. This opens an opportunity to utilise recycled carbon fibres in additive manufacturing. Among multiple additive manufacturing processes, fused deposition moulding (FDM) and selective laser sintering (SLS) are the two processes that can be used for manufacturing parts with recycled carbon fibres. It is vital that the recovered fibres are free of resin residue to be used in additive manufacturing [120]. Figure 13 shows the process by which additive manufacturing can be used to manufacture a part using recycled carbon fibre. In the FDM process, the recycled carbon fibres are grounded into chopped fibres using a ball mill. The chopped fibres are mixed with the desired powder (matrix) in certain ratios. The mixture is then extruded through a 3D printer, which can be used to manufacture the desired part [121].

Figure 13: Fibre remanufacturing using additive manufacturing [122]

The other method for additively manufacturing composite parts is by SLS process. In a SLS process a powerful laser is used to sinter the polymeric powders which is then used to create a structure with computer aided design [123]. This creates an opportunity to create parts using recycled fibres which can be used in automotive, marine and oil and gas industries [124].

6 TRACEABILITY OF CARBON FIBRE RECYCLING

The major problem experienced by the carbon fibre recycling industry is the traceability of carbon fibres. With end-of-life components it becomes harder to trace the origin of the component and the respective virgin carbon fibre used in them. In the current recycling chain, the only available traceability system is the use of dry fibre wastes to be reformatted into fabrics or prepregs [104]. The major barrier in developing a system that can aid the traceability of carbon fibres is that manufacturing and product development companies are not ready to share their intellectual property [125].

Recently, the French National Research Agency has funded project SEARRCH (Sustainability Engineering Assessment Research for Recycling Composites with High value) that is engaging with stakeholders from different groups such as CFRP manufacturers (which includes composite manufacturers, aircraft manufacturers and suppliers), end-of-life handlers (asset owners), institutional organisations (regulators and national agencies), and research laboratories. From this study, it was identified that recycled carbon fibres are not a viable alternative for manufacturing structural parts for an aircraft due to the lack of traceability and heterogeneity of the recycled fibres. This leads to certification issues and other related costs [126]. Development of an open loop system which aids the adoption of recycled composites to be used in other areas such as the automotive, energy and sports sectors may provide a solution. From the study, it was concluded that the diversity of resin and additives in an end-of-life component remain as a major challenge to tackle in carbon fibre recycling value chain. The proposed open loop system to address this issue is shown in Figure 14

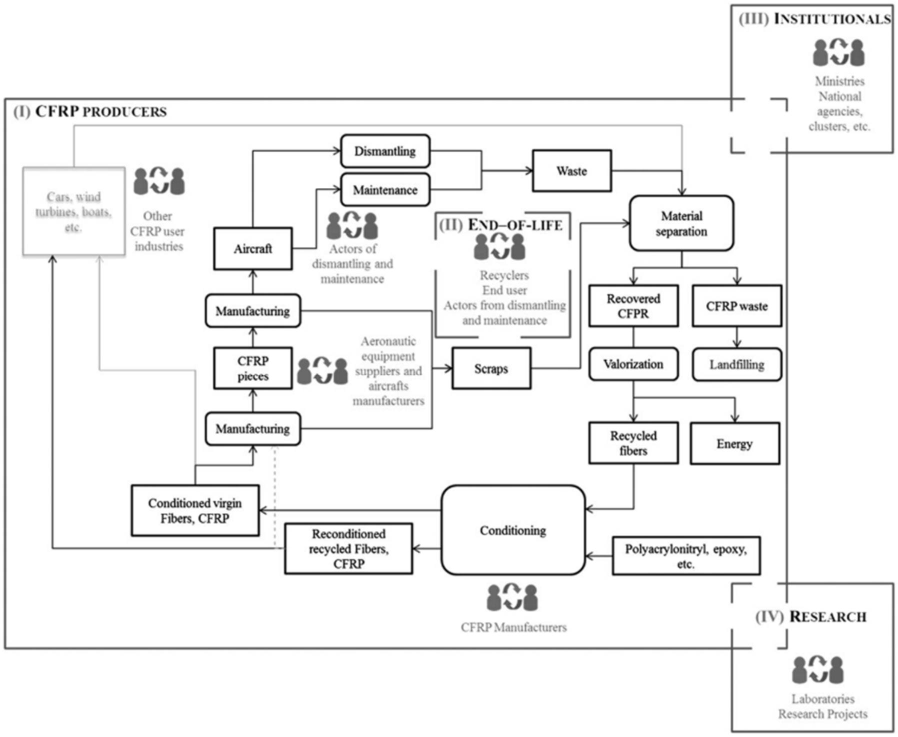

Figure 14: Open loop system for recycling carbon fibres from the SEARRCH project [126]

The findings of the SEARRCH study highlights the requirement of a database which can be used to track the essential information about the recyclable component and its constituents. Although, this may provide an ideal solution to the existing problem, it possesses issues such as data security and intellectual property. This leads to the requirement of a system which can be used to access the confidential information with security ensuring traceability of accessed data. The recent development in blockchain technology combined with Internet of Things (IoT) can ensure traceability of the data in the recycling chain with an extent of required visibility. Blockchain technology is widely used by cryptocurrencies such as bitcoin and Ethereum. In blockchain technology each transaction is recorded by a block. Each block in a blockchain has a unique hash value. Unique hash values are also created when these blocks are transacted. The hash values are tamperproof which makes the system secure [127]. Blockchain can be classified as private and public. In a public blockchain, anyone can join the network and be involved in contributing to the network. Whereas in a private blockchain the information is only shared with a restricted number of users within a group. The level of information shared between different members can also be restricted using smart contracts [128]. Smart contracts are legal contracts which are shared between the group members and which cannot be accessed by others who are not permitted to do so. Smart contracts contain all the necessary data such as certifications, transactions, and solid information [129].

Combining IoT technology such as RFID chips, or barcodes can be interlinked with blockchain which can provide access to immutable, transparent and private information [127]. By scanning the barcodes or RFID tags the recyclers can access required information for recycling and the origin of the product, recycling process, quality and safety measures for recycling [130]. Also, the recycler can add data back to the blockchain about the quality of the recycled material, encouraging traceability [131]. To implement blockchain technology in recycling Centobelli *et al.* [132] introduced a conceptual framework known as the triple retry framework as shown in Figure 15. The proposed triple retry framework is designed to interconnect three supply chain reverse process (recycle, redistribute, remanufacture) with three factors in the industry (trust, traceability, transparency) that affect the circular supply chain. The triple retry framework is based on trust between each participant in the recycling chain and they follow good practices to recycle the component. This trust can be backed by the smart contract in the blockchain which eliminates data manipulation. The blockchain ledger also provides traceability with each transaction being held in the ledger [132].

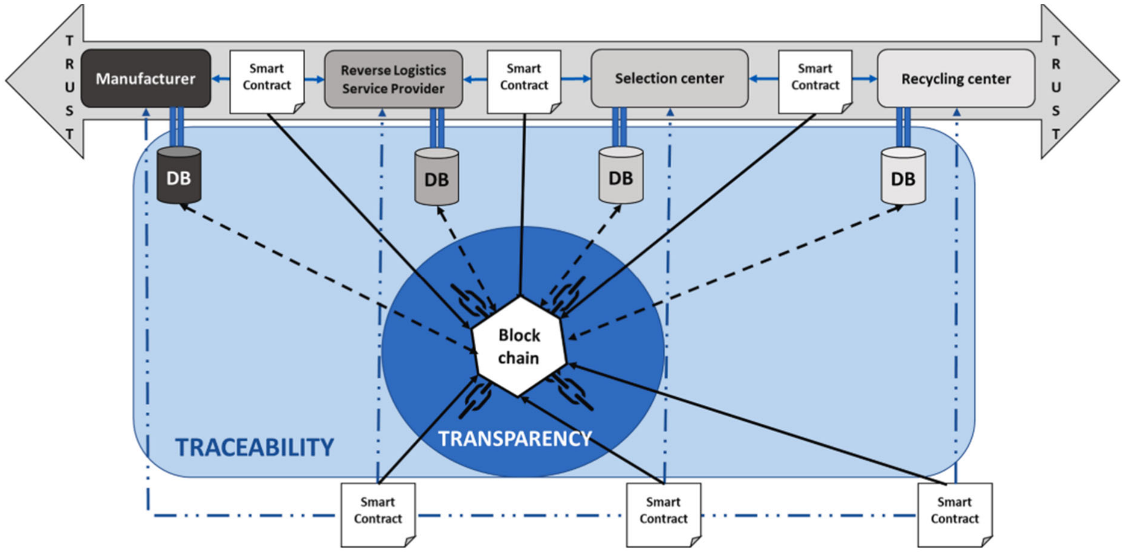

Figure 15: Triple retry Framework [132]

Although adapting blockchain technology to enhance material traceability and recycling has been investigated by other industries, there is no published literature about implementing blockchain in carbon fibre recycling. This indicates further development and exposure is required about this technology. Gong *et al.* lists some of the barriers in implementing this technology in the recycling industry. The barriers are widely classified as cognitive deficits, technology barriers, internal barriers and external barriers [131]. The cognitive deficits being limited recycling awareness and hesitation to adapt to technology such as blockchain. Technology defects leads to a lack of infrastructure and embedded systems. Internal barriers are caused by lack of capital resources and experience and external barriers can be due to a lack of government policies and industry support [131]. Developing relationships and working with multiple organisations involved in carbon fibres from manufacturing to recycling will help to implement such systems which can offer better traceability chain and better development of recycled composites.

7 CONCLUSION AND FUTURE WORKS

This study has reviewed the different methods of carbon fibre recovery and carbon fibre reformatting / remanufacturing. The fibre recovery methods from end-of-life CFRP components can be widely classified in to mechanical, thermal, chemical, and electrochemical based on the processes. Carbon fibres recovered from end-of life components following these recycling methods usually suffer significant damage and exhibit poor mechanical properties compared with virgin fibres. From this literature review it has been identified that there are no bespoke test standards for validating the mechanical properties of recycled fibres. As recycled fibres tend to vary a lot in fibre length, it is important to have an international standard that focuses on measuring the mechanical property of the recycled fibre.

Another huge gap that has been identified from this study is the traceability of recycled carbon fibres. Although significant progress has been made in the development of fibre recovery methods and repurposing, there is a huge gap in the literature with carbon fibre traceability and quality control. As the property of recycled fibre heavily depends on the precursor, a traceability chain or technology which can be used to track the source of the fibre and its properties must be established. This reiterates the necessity of material tracking systems and certification schemes for recycled carbon fibre. Having a material tracking system which can identify the source of the fibres using unique identifiers or labels will help to identify and track the fibres through various stages of the recycling process. Certifications from organisations like ISO or ASTM to categorise the recycled fibres will help to ensure that recycled fibres meet certain standards in terms of quality and the process of recycling. Transparency in terms of data and data generation is also very important as this will encourage organisations to make use of recycled fibres by increasing confidence in recycling processes.

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9 APPENDIX

9.1 INDUSTRIES INVOLVED IN COMPOSITE RECYCLING

Company	**Technology**	**Capacity (Tones/year)**
Adherent Technologies Inc. (USA) [133]	Pyrolysis	N/A
Alpha Recyclage Composites (France) [134]	Steam thermolysis	300
Carbon Conversions Inc. (Toyota Tsusho America, US) [135]	Pyrolysis	2000
CFK Valley Stade Recycling GmbH & Co. KG (Germany) [96]	Pyrolysis	1000
Curti SpA (Italy) [136]	Pyrolysis	12
Carbon Recycling Technology Centre (US) [137]	Recycling uncured prepregs	150
Gen 2 Carbon (Formerly ELG Carbon Fibre (UK) [138]	Pyrolysis	2000
Fairmat (France) [139]	Smart recycling (Using Artificial intelligence, robotics, computer vision, *etc*.)	5000
Hadeg Recycling Ltd. (Germany) [140]	Pyrolysis	N/A
Hitachi Chemical [141]	Solvolysis	12
IACMI (US) [142]	Pyrolysis	N/A
KARBOREK RCF (Italy) [143]	Pyrolysis	1000
Mixt Composite Recyclables (France) [144]	Grinding	N/A
Panasonic Electric Works (Japan) [145]	Hydrolysis	N/A
Procotex (Belgium) [136]	Mechanical	N/A
SGL Automotive Carbon Fibres (US) [136]	Pyrolysis	1500
Sigmatex (UK) [1]	Recycle dry fabric from production scrap	N/A
Takayasu [136]	Pyrolysis	60
Toray Industries [136]	Pyrolysis	1000
TRC (Spain) [1]	Pyrolysis	N/A
University of Manchester (UK) [1]	Mechanical	20
University of Nottingham (UK) [1]	Fluidised bed process pilot plant	100
V-Carbon (US) [1]	Solvolysis	1.7
Vartega inc (US) [146]	Mechanical	109