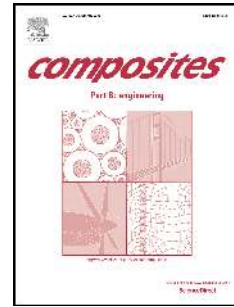


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Current Status of Carbon Fibre and Carbon Fibre Composites Recycling

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Abstract

The rapid rise in the applications of carbon fibre reinforced polymer matrix composites (CFRPs) is creating a waste recycling challenge. The use of high-performance thermoset polymers as the matrix makes the recovery of the fibres and the resins extremely difficult. Implementation of a circular economy that can eliminate waste and re-use resources warrants the use of efficient processes to recycle end-of-life CFRP components and manufacturing wastes. To this end, herein we present a critical review of the current technologies for recovering carbon fibres and/or the polymers and re-manufacturing CFRPs. New research opportunities in developing new biodegradable thermosets and thermoplastic matrices are also outlined together with more radical recycling strategies for the future.

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Keywords

A. Polymer-matrix composites (PMCs); B. Fibre/matrix bond; D. Mechanical testing; E. Recycling

1 Introduction

Carbon fibre reinforced polymer matrix composite (CFRP) materials are increasingly used in a wide range of industries, including air, land, and sea vehicles, wind turbines, storage tanks, and sports equipment. This soaring demand for CFRPs stems from their high specific strength, high specific stiffness, low density, ease of integration for part consolidation and multi-functionalities, and freedom in design. Driven by new targets of lower CO₂ emissions and the need for light-weighting of vehicular structures, the global demand for composites in aerospace, construction, wind energy and automotive is projected to continue to grow at a compound annual growth rate (CAGR) of approximately 6%, with the total market increasing from \$26 billion in 2018 to \$41.4 billion in 2025 [1, 2]. The global demand for carbon fibre is expected to reach 117 kilo tonnes (kt), and 194 kt for carbon fibre reinforced polymer or plastic (CFRP) in 2022, which represents a CAGR of around 11.50% and 11.98% through the year 2010 [3]. The rapid growth in the adoption of CFRPs in existing and emerging industry sectors presents major environmental challenges in wastes, such as off-cuts generated during composite manufacturing (as much as 40%), and end-of-life CFRP products. The global CFRP waste is projected to increase to 20 kt annually by 2025 [4]. With around 6000–8000 commercial aircraft reaching their end-of-life by 2030 [5], there is a clear and exigent demand for developing economically-sustainable waste management and recycling techniques for CFRPs.

The production of carbon fibres is an energy-intensive process that consumes around 198–595 MJ/kg [2]. As a result, carbon fibres possess higher (approx. 10 times) embodied energies than other synthetic fibres, such as glass fibres. Recycling the used carbon fibres can hence significantly reduce their potential environmental impact. According to the recent literature, 38.4 MJ/kg is consumed to recycle CFRP by a chemical method [6], which is close to 10~30 % of the total energy required to manufacture virgin fibres.

CFRP recycling is often dictated by the properties of the polymer matrix. Currently, thermoset polymers (e.g., epoxy and unsaturated polyester) are mainly used as matrices for structural CFRPs, representing around 80% of polymer matrix composites (PMCs) [7], attributed to their high mechanical strength, high chemical and thermal resistance, dimensional stability, and durability. However, the irreversible cross-linked structure of cured thermoset polymers leads to significant difficulties in the recovery of the fibres and the thermosetting polymer, since they, once cured, cannot be melted, remoulded, reprocessed, or re-crosslinked into solid polymers [8]. The chemical degradation of cross-linked thermoset networks occurs under aggressive chemical or high-temperature reaction conditions, which can also cause damage to the recovered carbon fibres.

Researchers and industry are currently working towards a *circular economy*, which is an industrial system aimed at reusing, remanufacturing, and recycling of products at their end-of-life (EOL) stage. When reuse and remanufacturing, and further reselling of products is impracticable, closed-loop recycling is exercised to retain materials in use and circulation [9]. Recycling within the circular economy is particularly deemed effective when the physical characteristics of the material are conducive for easy transformation. Overall, the circular economy aims at using renewable energy, eliminating toxic chemicals, and eliminating waste by better design of materials, products, systems and business models [10]. This strategy has been reflected in some recently published works [11, 12] on zero-waste manufacturing.

This review aims to provide a comprehensive evaluation of the state-of-the-art in the recycling of carbon fibres and CFRPs, and the current and future applications of recovered carbon fibres (RCFs). Within this review, the concepts of the circular economy pertinent to the recycling of CFRPs are highlighted in terms of EOL CFRP transformation and manufacturing wastes to RCFs, recycled polymers, and re-manufactured CFRPs. The development of recyclable thermosets, broader use of thermoplastics matrices, and utilisation of low-cost methods for separating thermoset matrix from carbon fibre reinforcement will also be discussed.

2 Carbon fibre composites market and recycling challenges

The global demand for CFRP is growing steadily in both the established and new sectors, causing challenges for recycling and managing waste.

2.1 Global market for carbon fibre composites

The demand for CFRPs has tripled from 2010 to 2020 (**Fig. 1**) and is expected to surpass 190 kt by the year 2050 [13-15]. The global CFRP demand in 2018 (**Fig. 2a**) shows predominant CFRP use in aerospace, wind turbine blade, sport and leisure, and automotive applications. Interestingly, the implementation of CFRPs in wind turbine blade manufacturing has driven a higher volume (34.2 kt) utilisation, exceeding their usage in traditional aircraft (32.8 kt) applications. CFRP pressure vessel applications for gas and liquid storage constitute 8% or 11.4 kt of global demand. However, when analysed in terms of cost, the sector-wise distribution of CFRP leans towards the aerospace sector, which constitutes 73% of CFRP sales (**Fig. 2b**). Sport and leisure commercial products amount to 7% of the revenue, exceeding CFRP usage in the automotive sector (6%). The fraction of CFRP sales in the wind turbine and automotive sectors are relatively low, owing to the usage of low-cost carbon fibres.

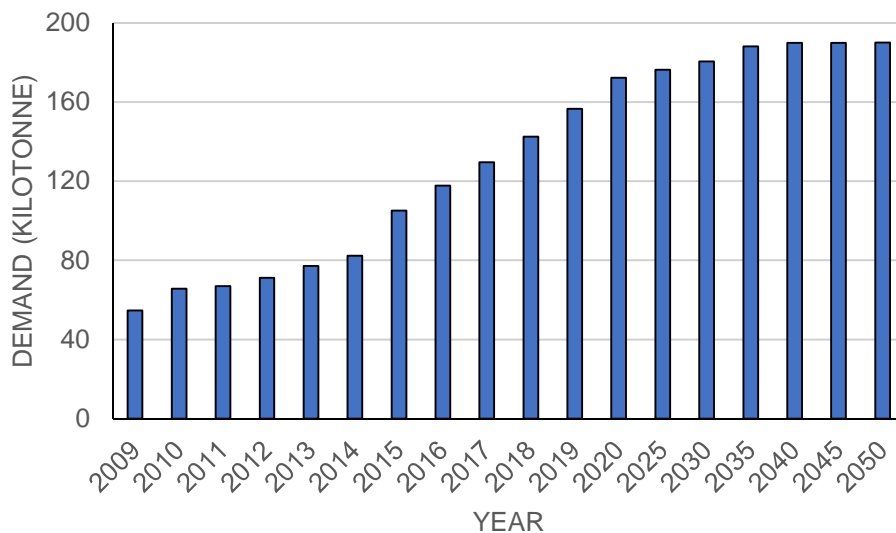


Fig. 1. Global CFRP demand in kilo tonnes by year [16, 17].

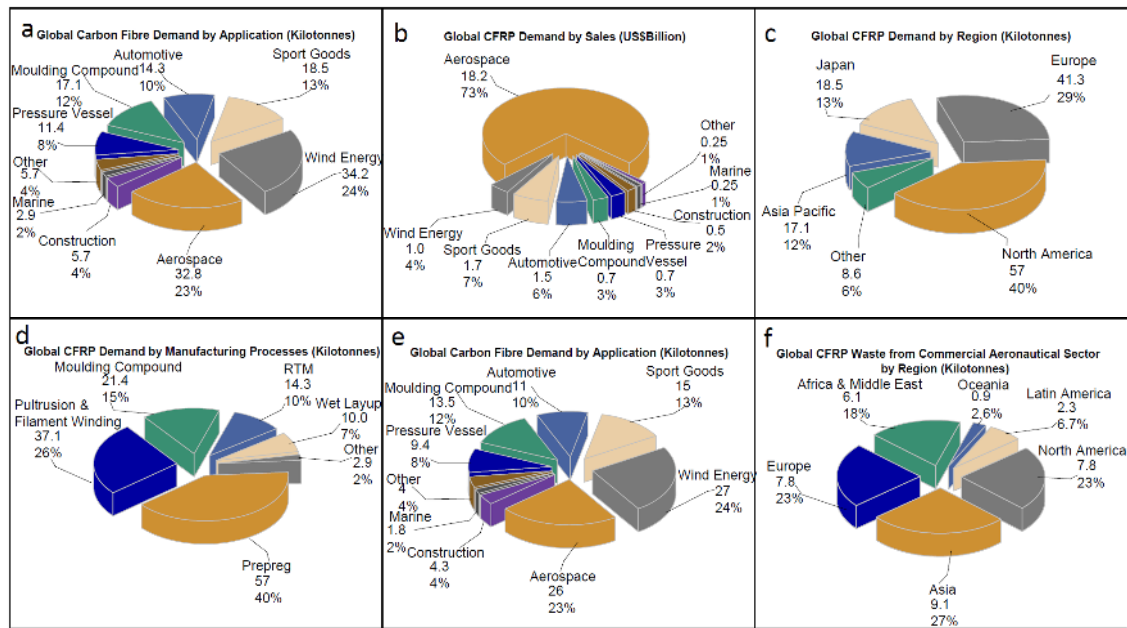


Fig. 2. Global CFRP demand in the Year 2018 classified by (a) application, (b) sales, (c) region, and (d) manufacturing process. The global carbon fibre demand in 2020 (e) by application, and (f) estimated Global CFRP waste in 2050 from the aeronautical sector by region [16, 18, 19].

North America, Europe, Japan, and Asia Pacific regions constitute the majority of demand for CFRPs (**Fig. 2c**). North America and Europe used 40% and 29% of the CFRP composites, while Japan and the rest of the Asia Pacific accounted for a combined 25% of CFRPs. Boeing, Airbus, defence, and other automotive and wind energy companies in North America and Europe led CFRP sales in these two regions. The CFRP use was significantly lower in Japan compared to the US and Europe, although the highest quality carbon fibres are produced in Japan.

The global CFRP market is classified according to the composite manufacturing methods shown in **Fig. 2d**. The conventional prepreg and lay-up processing are still the most widely used routes for manufacturing products in aerospace and sport (40%) applications. Pultrusion and filament winding are prevalent in wind turbine blade fabrication and rotary part construction, accounting for 26% of the share. Moulding compounds based on discontinuous carbon fibre reinforced polymer composites such as sheet moulding compound (SMC) are prevalent in automotive applications. Out-of-

autoclave methods such as resin transfer moulding (RTM) and its variations, especially high-pressure RTM, is gaining acceptance in automotive production because of its potential to reduce cycle times to 10 min, albeit an associated high tooling cost that inhibits its progression in industrial adoption [20].

The future market for CFRP composites encompasses aerospace and aircraft, sport and leisure, wind blade, automotive, pressure vessel, moulding compound, construction, and marine applications. The estimated carbon fibre demand for aerospace use is 26 kt in 2020 (**Fig. 2e**), which is predominately (70%) driven by commercial aircraft manufacturing (e.g., Boeing 787 and Airbus A 350). The estimated carbon fibre demand for use in sport and leisure is 15 kt in 2020, mainly in golfing, fishing, bicycles, racquets, hockey, and ski poles. The increased use of carbon fibres in bicycles is the main driver in this sector. A comparable amount of carbon fibres to aerospace are used by building wind blades largely because of the decrease in the cost of carbon fibres, and the availability of novel manufacturing processes that enable the fabrication of large-diameter wind turbines. Today, the CFRP industry is witnessing a shift with Vestas, Nortex, and GE promoting the broader use of lightweight carbon fibres.

The automotive market is driven by cost reduction strategies encompassing reduction of cycle time and lowering of material cost. A synergy of low-cost carbon fibres for reducing material cost, and innovative design enabling the utilisation of lightweight carbon fibres is required to drive this change in the automotive industry. The hydrogen economy, particularly with the gas and liquid storage pressure vessels are increasingly built by CFRP, and this demand is projected to rise with novel fuel cell vehicles and hydrogen transportation and storage becoming more commonplace.

Discontinuous fibre-reinforced composites feature short and long carbon fibre reinforcements, with fibre aspect ratio (l/d) lower than 10,000. These discontinuous carbon fibre composites can achieve a high fraction of mechanical performance at lower material cost, higher production rate, and therefore realising the balance between component quality and economic cost, in comparison with continuous CFRP composites. CFRP composites are being increasingly used for reinforcing old buildings and to construct offshore structures such as oil platforms because of the high corrosion

resistance of CFRPs. Also, CFRPs are expected to gain prominence in building racing boats, luxury cruises, military ships, increasing carbon fibre utilisation from 0.8 kt in 2010 to approx. 1.8 kt in 2020. Emerging markets for CFRPs, such as electronics, are adopting CFRP composite thin laminates and short carbon fibre plastics for lightweight and aesthetics. Furthermore, high-speed trains and electrode material markets are using more carbon fibre materials such as hybrid composites and carbon fibre papers.

2.2 Recycling challenges of CFRP waste

Global growth in CFRP utilisation has brought about a concomitant increase in production waste (e.g, off-cuts) and EOL components (e.g., decommissioned aircraft), which will grow correspondingly with the number of aircraft reaching their EOL. **Fig. 2f** shows the estimated CFRP waste in the commercial aviation sector from production waste and EOL aircraft by region in 2050. The annual waste to be processed will reach estimated 34.2 kt in 2050, with the highest cumulative growth concentrated in Europe, North America, and Asia [21]. According to a review of composite waste in the UK supply chain in 2015, nearly 98% of the EOL components and manufacturing composite waste were buried in landfills [22]. Landfill and incineration recovery of the embodied energy [23, 24] are two traditional waste treatment methods, with a majority of FRP wastes currently being buried in landfill sites. The energy generated from incineration is approximately 30 MJ/kg for CFRP waste [25] and the potential of recovered energy depends on the energy content of the materials and efficiency of the incinerator [2]. However, both incineration and landfilling are not classified as recycling since they do not involve a recovery operation by which waste materials are reprocessed into products, materials or substances whether for the original or alternate purposes [26].

The 2016-2017 rate of the landfill in the UK was £84.40 per tonne, and the cost of the landfill included transport and gate fees of £120~130 per tonne. The European Commission put forth a target of reducing municipal waste to 10% by 2030, with landfills banned in some European countries. Moreover, the European Waste Framework Directive (2008/98/EC) implemented a “polluter pays” principle, stating “by 2020 a minimum of 70% (by weight) of non-hazardous construction and demolition waste... shall be prepared for re-use, recycled or undergo another material recovery”

[27]. The End-of-Life Vehicle Directive (ELV, 2000/53/EC) set targets that require a minimum of 95% of a new vehicle by average weight to be recycled at the EOL stage, and 85% of the waste to be re-used by the European Commission [23]. Similarly, the Waste Electrical and Electronic Equipment Directive (WEEE, 2012/19/EU) set targets for collection, recycling and recovery of electrical components. These increasingly stringent legislations cause changes to how waste is created and managed within the operations and supply chains of the composites industry and compel manufacturers to seek novel, alternative, and more efficient recycling routes for CFRP wastes.

3 Current thermoset matrix composites recycling routes

CFRP recycling can be categorised into three broad methods, i.e., mechanical, thermal, chemical (Fig. 3), which uses a corresponding energy source to separate the carbon fibres from the thermoset matrix respectively.

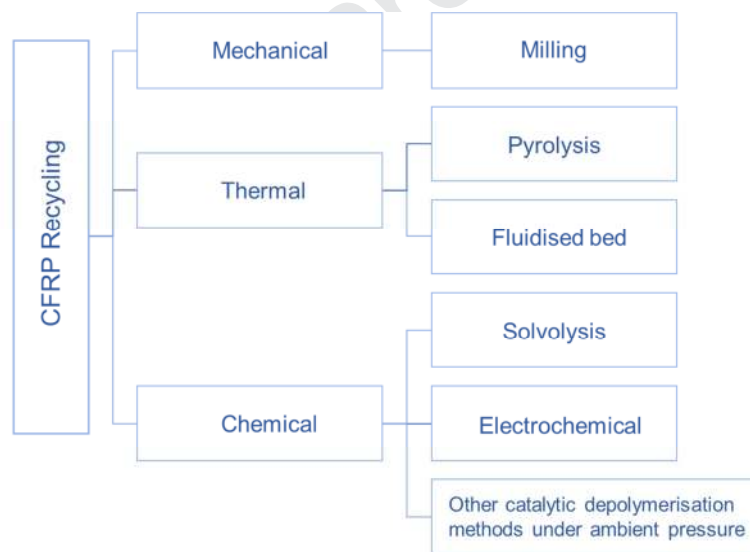


Fig. 3. A classification of the major CFRP recycling methods.

3.1 Mechanical recycling-milling

Mechanical recycling is technologically the most mature CFRP recycling method, which involves multiple steps to reduce the size of the waste. Firstly, composites are pulled apart and shredded to a 50~100 mm size, and further grinding or milling is applied to obtain recyclates with different sizes that range from fine powder to fibrous

recyclates [2, 28]. **Fig. 4** shows a common granulator (Wittmann MAS1) with a processing capacity of 30 kg/h, with a fixed drum speed of 200 rpm, and an exit sieve aperture diameter of 5 mm [29], which allows powder and fibrous recyclates to be collected in a bin. The recyclates from this mechanical recycling can be used as fillers in short fibre composites such as sheet moulding compounds (SMCs). Because of the resultant short fibre aspect ratio, these recyclates possess a relatively low market value.

Because of the attrition caused by CFRPs during recycling, the ensuing damage to the processing equipment often increases the operational cost of mechanical recycling, hence decreasing the economic margin of the recyclates, and often making this a questionable choice in terms of viability. The mechanical milling process is albeit nontoxic, and the process can be performed at room temperature [30]. This process can accommodate waste of size up to 50 mm, and attain 50% to 65% residual tensile strength of the virgin carbon fibre strength. The main health and safety hazard for this recycling method is the dust from the recycling system, which is precluded with proper engineering controls and use of personal protective equipment in addition to well-ventilated work areas with particulate and dust extraction.

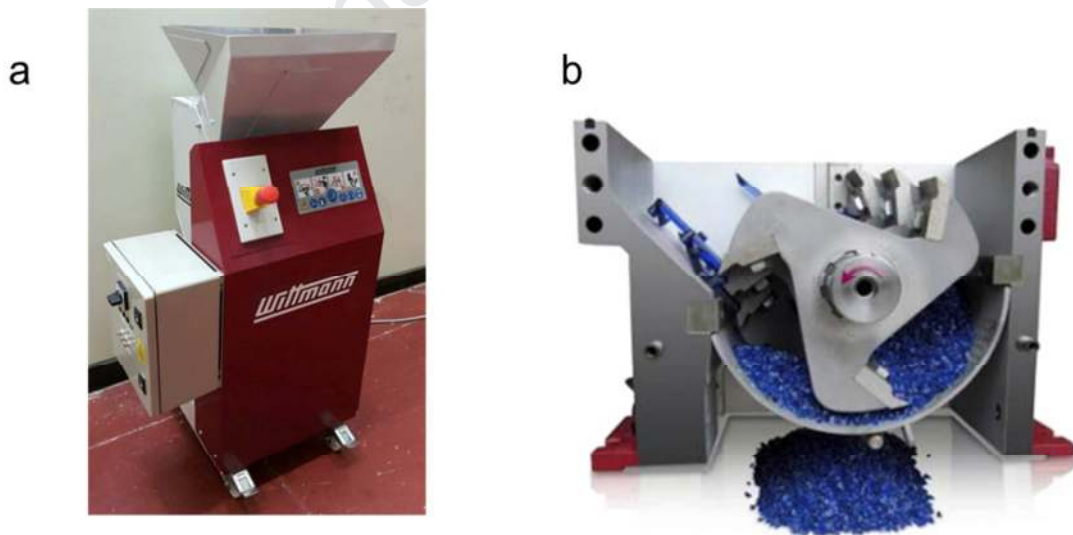


Fig. 4. The (a) MAS1 Wittmann granulator and its rotor blades and (b) cutting chamber [29].

3.2 Thermal recycling

Thermal recycling methods for CFRP incorporate the action of high temperature and fluids to break down the matrix fibre and uncover the fibres and are classified as pyrolysis and fluidized bed recycling or the variants thereof.

3.2.1 Pyrolysis

Pyrolysis is a thermal decomposition process performed in an inert gas atmosphere without oxygen where the thermoset composite is heated to a temperature between 400~1000 °C enabling recovery of long carbon fibres with high modulus [2], which also exhibit 50-85% of the tensile strength of virgin carbon fibres [30]. During pyrolysis, the decomposed and de-constituted resin matrix is removed from the fibre reinforcement, generating gas, oil, tar, and char [31, 32]. Post-pyrolysis of the recyclate in the air is required to remove the char residue and obtain clean fibres for reuse. Usually, polyester resins require a lower temperature for complete conversion, but epoxy resins require a higher temperature for decomposition. The selection criteria for processing temperature is dictated by the degree of conversion of resin that precludes significant mechanical property loss of the reinforcement fibres. Giorgini et al. investigated the pyrolysis process for fibre reinforced polyester composites scraps in their 70 kg pilot plant, and found that pyrolysis at 500~600 °C yielded ~20 wt% of oil, ~40 wt% of gas, and ~40 wt% solid residue. Further oxidation treatment at 500 °C for 50 min or at 600 °C for 20 min was sufficient to obtain clean fibres [33]. A novel super-heated-steam method (at 550 °C) was used to reclaim high-quality carbon fibres with no char residue on the fibre surface [34].

A recent review of using pyrolysis to recycle EOL CFRPs [35] with a critique of pyrolysis processing conditions of composite wastes, the associated technical challenges, and re-use applications for the recycled fibres. The pyrolysis process was identified as the most viable and sustainable CFRP recycling process to achieve process and resource efficiency.

3.2.2 Fluidized bed recycling

Fluidized bed recycling (**Fig. 5**) is under development at the University of Nottingham since the 2000s and it is now operational at a pilot-scale [36]. After shredding to typically 6~20 mm [6], the composite waste is fed into a bed of silica sand (with size <1 mm) which is fluidised by the stream of hot air at a temperature between 450 and 550 °C, at pressures of 10 – 25 kPa [30]. The main work health hazards of this process are the pollutant gas, presence of organic solvents, and the usage of high energy, which guides the selection criteria of the recycling conditions. The reaction temperature is selected to ensure decomposition of thermoset matrix without significant fibre degradation. Upon rapid heating, the fibres are released by attrition and the thermal decomposition and degradation of the matrix, and then separated and collected. The gases released from the matrix are subjected to combustion to oxidize the by-products. The recycled fibre length is 5~10 mm, and a corresponding 10-75% retention of the tensile strength of virgin carbon fibres can be achieved [30, 37]. This process is suitable for EOL composite components since rivets, bolts and other fittings can be collected in the bed, and carbon fibres from composites can be recycled [2]. Overall, the reuse of fluidised bed RCFs is competitive in terms of their limited environmental impact [6].

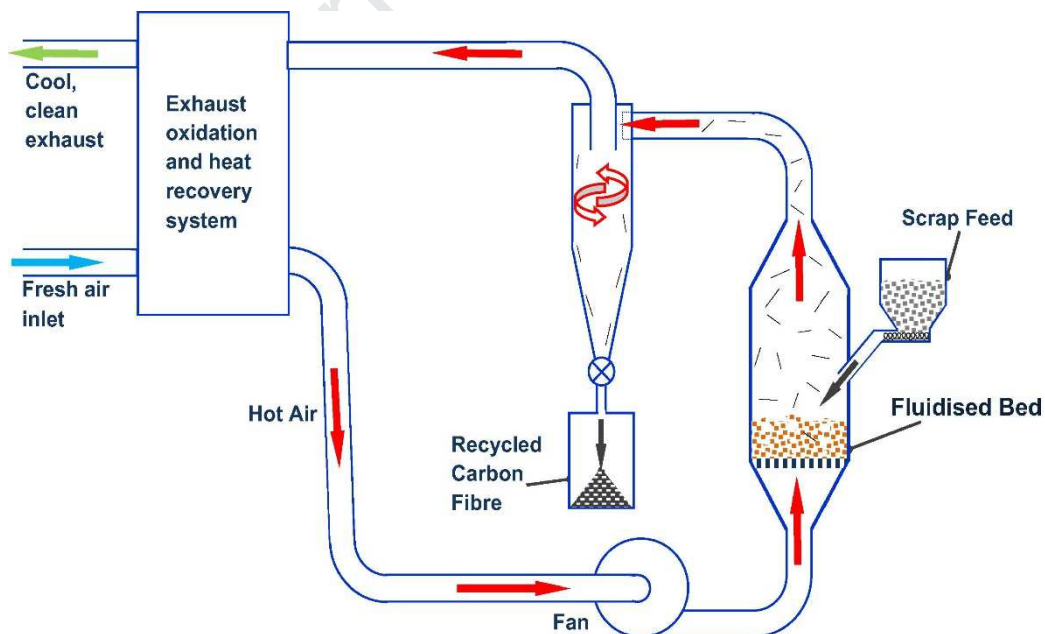


Fig. 5. A schematic of the configuration and flow of the fluidised bed CFRP recycling process. Reprinted from [6], Copyright (2019), with permission from Elsevier.

3.3 Chemical recycling

Chemical and associated electrochemical methods of CFRP recycling use a divergent range of solvents, both supercritical and subcritical, and often are operated at ambient pressures. The electrochemical recycling methods use high electrical currents to decompose the CFRP constituents, whereas catalytic depolymerisation methods also are efficient at ambient pressure to achieve a high degree of clean, residue-free carbon fibres.

3.3.1 Solvolysis using supercritical/subcritical solvent

Solvolysis uses solvents or heated solvents to decompose thermoset matrix to separate fibres from the matrix. Solvolysis offers many ways of recycling because of the availability of a broad range of solvents combined with different processing variables such as temperature and pressure, and often in conjunction with a catalyst. The chemical processing is usually classified into supercritical, subcritical, and near-critical [2].

Water is widely used over other solvents due to its environmentally friendly nature. Other solvents such as ethanol, acetone, methanol and their mixture with water at low critical temperature and pressure have also been studied [38]. The chemical recycling process can produce long RCFs with no residue on the surface, therefore, high-quality carbon fibres with a minor reduction in mechanical properties can be obtained. Compared with pyrolysis, the chemical treatment uses a lower temperature for processing and generates cleaner and longer recycled fibres, however, solvolysis requires expensive equipment that is ought to resist corrosion at elevated temperatures and pressures and bears a negative impact on the environment and human physiology. These characteristics of the fluidized bed process often limit its industrial-scale application [39].

Phenol and potassium hydroxide (KOH) were used to chemically recycle CFRP by Liu et al., who found an optimal ratio of KOH mass/phenol mass/water volume (1/10/100, g/g/mL) exhibiting the highest decomposition efficiency with no tensile property loss in the recovered carbon fibres [40]. Liu et al. depolymerised cured epoxy resin in subcritical methanol in the presence of potassium hydroxide (KOH) at a

relatively low reaction temperature (210°C), and found that with 0.036 mol L⁻¹ KOH catalyst, the complete depolymerisation temperature decreased to 65°C [41]. Zhang et al. reported a degradation method using an organocatalyst, N-methyl-4-piperidinol, in ethylene glycol to depolymerise methylcyclohexene-1,2-dicarboxylic anhydride (MeTHPA)-cured epoxy resin [42]. The catalyst could be recovered by vacuum distillation, in contrast to alkaline catalysts that are usually difficult to remove from degradation products.

Kuang et al. recycled EPON 828 epoxy, which is a common bisphenol A diglycidyl ether resin in the composite industry, using a solvent mixture at relatively low pressure and mild temperatures [43]. The anhydride-epoxy network was depolymerized by the selective ester bond cleavage process via a solvent-assisted, alcohol-assisted transesterification reaction. At 170 °C in the atmosphere, the thermoset was dissolved by 50% in 28 min, and by 95% in 70 min. They were also able to reclaim carbon fibres (**Fig. 6**) under the same conditions in 90 min and use the decomposed epoxy oligomer to synthesize new epoxies with high mechanical properties. Kim et al. used supercritical fluid water to recycle TORAYCA T700 carbon fibres from commercial composites with 99.5% epoxy matrix removed by optimising treatment conditions [44]. The CFRP composite was immersed in 0.30 M TBD-EG/NMP (10/90) solution (150 mL) and heated at 170 °C for 1.5 h to dissolve the epoxy binder. The carbon fabric was reclaimed after washing with acetone and subsequent drying.

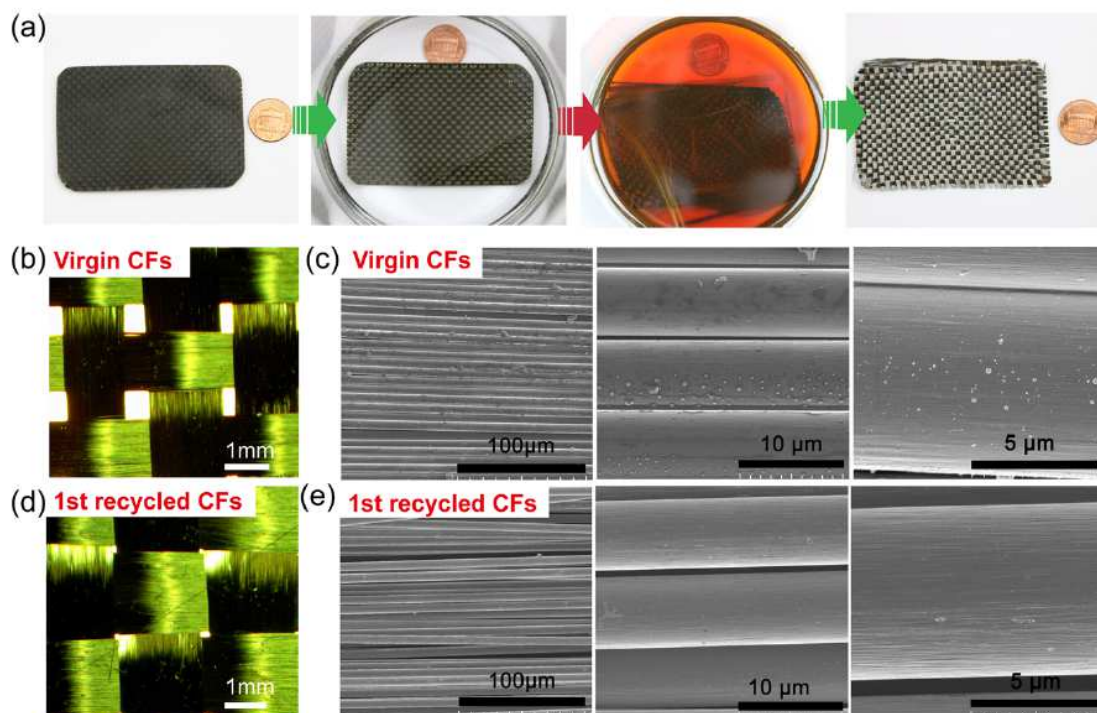


Fig. 6. Recovery and reuse of carbon fibres from CFRP, showing (a) recycling of composites laminate, and a comparison of optical microscopy images and SEM micrographs of virgin carbon fibres ((b) and (c)) and RCFs ((d) and (e)) elucidating indiscernible differences. Reprinted with permission from [43]. Copyright (2019) American Chemical Society.

3.3.2 Solvolysis using solvent at ambient pressure

Solvolysis at ambient pressure possesses unique advantages in limiting damage to the RCFs. Liu et al. utilised a mild chemical recycling method with $ZnCl_2$ /ethanol catalyst to recycle CFRP with high T_g (>200 °C) [45]. The low temperature of treatment (<200 °C) caused negligible damage to the RCFs, and the reclaimed polymer matrix was shown to retain high strength and modulus after adding to other new epoxies in loadings up to 15 wt%. A catalytic oxidation method was demonstrated to recycle the most thermally stable benzoxazines under mild conditions. The thermoset matrix was catalytically depolymerized through hydride abstraction with a ruthenium catalyst [46].

Hou's group has developed an effective chemical recycling method by using $AlCl_3/CH_3COOH$ as a catalyst to realise selective cleavage of the tertiary carbon-

nitrogen bond [47]. By applying the optimal recycling conditions (catalyst mass fraction of 15wt%, 180°C, 6h), up to 97% of the cured epoxy matrix was recovered. The recovered carbon fibres retained 98% of the tensile strength of the virgin carbon fibre reinforcement. Supercritical 1-propanol to recycle CFRP was also investigated, where 1 wt% of KOH additive in 1-propanol showed enhanced recovery efficiency with slight mechanical property loss [48].

3.3.3 Electrochemical recycling

The high voltage fragmentation method of electrochemical recycling uses pulsed electrical discharges to disintegrate solid material, which was used previously to fragment rocks to obtain valuable ingredients in mining. **Fig. 7** shows the mechanism of the process, indicating the temperature increase of the vessel after processing and the energy consumption at different stages [49]. A high voltage electrical pulse is generated between the electrodes and transports through the materials in the vessel. High energy consumption has been the biggest limiting factor since 75~80% of the energy is introduced into the processing vessel instead of the waste materials. Research has been conducted to compare mechanical recycling with high voltage fragmentation methods [50]. This method produces longer and cleaner recycled fibres than mechanical recycling, however, the energy consumed is 2.6 times more than that used for mechanical processing. Despite claims of a positive influence of high voltage fragmentation on separating composite constituents [51, 52], Leibner et al. presented a comprehensive evaluation of the practicality of high voltage fragmentation method as an alternative to mechanical comminution and found that this method lacks cost competitiveness compared with mechanical comminution, beyond the laboratory scale [49]. Zhu et al. used electrochemical recycling method on CFRP composites, which were immersed in a NaCl electrolyte containing the KOH catalyst in the presence of electrical currents [30]. By exerting the electrically-driven catalytic reaction, the C–N bonds of the epoxy resin were cleaved, leading to matrix decomposition through the electrochemical promotion of catalysis effect. Conventional, non-toxic chemicals were used under ambient temperature and pressure in this method, which realised near 100% resin removal and the retention rate of tensile strength was 90% for the recovered carbon fibres.

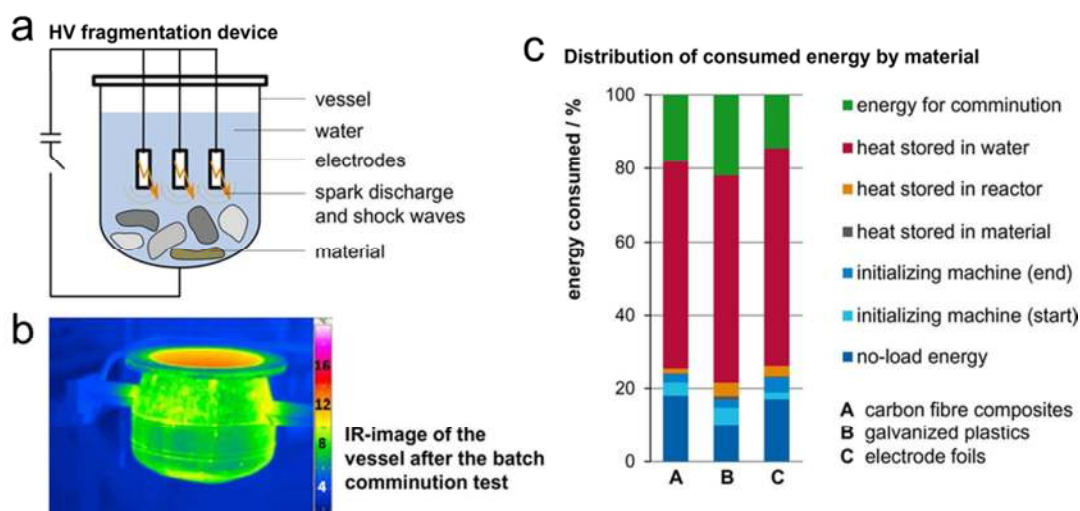


Fig. 7. A pictorial collage of high voltage fragmentation method showing a) schematic of the process, b) infrared image post-processing, and c) distribution of energy consumed by different materials. Reprinted from [49], Copyright (2019), with permission from Elsevier.

3.3.4 Other ambient-pressure catalytic depolymerisation methods

Molten alkali or salt are effective reagents that can process under mild conditions at atmospheric pressure. CFRP was decomposed in molten KOH, and the depolymerisation was carried out at a temperature range of 285~330°C, under ambient pressure [53]. The residual tensile strength of the recovered carbon fibres was >95% of the virgin fibres, with the process being particularly effective in removing contaminants in composite waste including thermoplastic paint, and sealants. The use of molten salt reduced the pyrolysis temperature and the energy consumption for epoxy. Wu et al. studied the catalytic pyrolysis of T700 carbon fibre/epoxy composites in molten salt $ZnCl_2$ and concluded that the epoxy composites can be completely degraded at 360 °C in 80 min under standard pressure to yield reclaimed carbon fibres with a high strength retention rate (~95%) of the virgin fibres [54].

4 Commercial CFRP recycling technologies and recovered carbon fibres

A diverse range of CFRP recycling vendors operate in different parts of the world, and specialize in a variety of aforementioned recycling technologies, expectedly

bringing about variability to the recycling market overall. In this section, the current state of major vendors, and the properties and the economic perspectives of the recovered carbon fibres from novel circular economy methodologies are explored in detail.

4.1 Current market players of CFRP recycling

CFRP recycling companies have been founded in the past twenty years from different countries in Europe, the USA, and Japan [2, 55]. **Table 1** lists the major companies and their commercial processing and production technologies. Pyrolysis process is currently predominately used for carbon fibre recovery, as it is well developed and can be applied at different stages of composite manufacturing and is commercially viable. The pyrolysis process for carbon fibre has a technology readiness level (TRL) of 8, which indicates the technology is currently at its “system/subsystem development” stage. By contrast, solvolysis technologies are commercialised to produce higher quality RCFs, using lower energy and lower environmental impact. However, a profit margin is difficult to achieve using such a chemical recycling method, despite the elimination of pre-treatment of CFRPs and the application of normal pressure during processing [56]. Solvolysis possesses a TRL 4, which is at its “technology development” stage [57]. Factors that drive the FRP recycling include the rising cost of landfills, the government and legislation restrictions and the social awareness of the importance of circular economy. However, above all, the most critical factor is new market penetration that makes CFRP recycling commercially viable. Examples of efforts towards this can be demonstrated by the formation of the first material supply chain between ELG and Boeing in the beginning of 2019, indicating the deployment of recycled CFRP composite materials in the aerospace industry. The current increasing adoption of recycled composite materials in automobiles by BMW and Volkswagen promotes the economic viability of recycled CFRPs in the automotive industry. With the pressing need to create higher-value recycling routes, diverse recovered carbon fibre products (e.g., nonwoven mats, aligned short fibre mats, and carbon fibre yarns) and recycled composite products (compounded thermoplastic pellets for injection moulding, 3D printing filaments, discontinuous fibre-reinforced thermoplastic sheet, etc.) are being developed [58]. For example, Shocker Composites and R&M International have

collaborated to produce densified recovered carbon fibres and thermoplastic/recovered CF pellets. The large-scale 3D printing results from using these recycled materials have shown to be similar to virgin CF products, albeit achievable at a lower cost. The new recycled materials with customized component designs offer new opportunities for economical recycled products.

Table 1. Current CFRP composite recycling companies in production and their technologies

Recycling Company	Recycling Technology	Processing Capacity (ton/year)
Alpha Recyclage Composites (France)	Steam thermolysis process	300
Carbon Conversions	Pyrolysis	2000
CFK Valley Stade Recycling GmbH & Co. KG (Germany)	Pyrolysis	1000
Composite Recycling Technology Centre	Recycle scrap carbon fibres and Uncured prepregs	150
ELG Carbon Fibre (UK)	Pyrolysis	2000
Hitachi Chemical	Solvolytic	12
IACMI	Controlled pyrolysis process	N/A
KARBOREK RCF (Italy)	Pyrolysis (reclaim both carbon and glass fibres)	1000
Pilot plant at University of Nottingham (UK)	Fluidized bed	100
SGL Automotive Carbon Fibres US site	Pyrolysis which uses released gas as an energy source for carbon fibre recovering	1500
Sigmatex (UK)	Recycle dry carbon fibre production scrap into the non-crimp fabric	N/A
Takayasu	Pyrolysis	60
Toray Industries	Pyrolysis	1000
TRC (Spain)	Pyrolysis	N/A
University of Manchester (UK)	Mechanical	~20 (calculated from lab capacity)
V-Carbon US site	Low pressure, moderate temperature Solvolytic	1.7

4.2 Property of recovered carbon fibres

Carbon fibres reclaimed by different CFRP recycling routes exhibit different retention rates of mechanical properties, with respect to virgin carbon fibres. **Table 2** displays the large variation among RCFs recovered by pyrolysis methods. The chemical recycling methods, however, produce RCFs with high retention rates because of the cleaner fibre surface after chemical treatment, which contrasts with the pyrolysis

recyclate, which leaves resin residues on the fibre surface. After the fluid-bed pyrolysis, the carbon fibres show around 20% loss in tensile strength due to the high temperature and attrition by the fluidised sand.

A range of RCFs is now available commercially, with a divergent range of sizing and mechanical properties (**Table 3**). Chopped mat, discontinuous short and long fibres and nonwovens are the most common fibre architectures available after post-processing. Often, RCFs are combined with virgin carbon fibres to compensate for the loss in mechanical and physical properties within the recovered fibres. The sizing content also differs as a function of the recycling method used, i.e., superheated steam processes aid in retaining the sizing, whereas thermal processes often lead to the loss of fibre sizing.

Table 2. The retention rate of tensile properties of recovered carbon fibres by different recycling processes

Recycling technology	Virgin fibres	Retention Rate of Tensile Strength of Recycled Carbon Fibre (%)
Pyrolysis	AS4-3K	15–98 [59]
Steam thermolysis	AS4C	95-99[60]
Solvolysis	TohoTenax C124	97-98[61]
Supercritical water	Hexcel 48192 C 1270 ST	94-98[62]
Fluid-bed	Toray T800	82[63]

Table 3. Commercially available as-received RCF properties

Property	Unit	Carbiso Milled	Carbiso CT	Carbiso CT+	Carbiso Chopped Carbon Fibre	Zoltek Type 45/65
Bulk density	g/l	400	-	-	-	350
Carbon fibre content	%	>95	100	100	>95	
Fibre density	kg/m ³		1800	1800	1800	-
Fibre diameter	Avg., µm	7	7	7	-	-
Fibre length	Avg., µm	80 to 100	6 and 12 (±1)	6 and 12 (±1)	3- 10 10-30	6
Other fibre	%	<5	0	-	60-90 <5	-

content						
Sizing content	%	0	<1.5	<5	-	2.5
Tensile strength	MPa	4150	5000	4150	3470	150
Tensile Modulus	GPa	230-255	262-292	230-255	246	15

4.3 Financial viability of recycled carbon fibres

In a recent report on cost analysis and market opportunities for RCF in the automotive industry, researchers found that the recovery of carbon fibres can be achieved at \$5/kg or less, based on a range of process parameters including plant capacity of 500 t/yr.- 6000 t/yr., which accounts for approximately 15% of the cost of virgin carbon fibre [64]. For smaller production capacity of 100 t/yr., RCFs have to realise a market value of \$15/kg to be financially viable. Their study also elucidates that recycled CFRP can provide significant weight savings versus steel and possess comparable mechanical properties at a comparable cost. In particular, the aligned recycled CFRP can greatly improve financial competitiveness (>35% cost reduction) because of the higher mechanical properties, with lighter structures. Another economic assessment shows that there is a high potential for RCFs to replace virgin carbon fibres or virgin glass fibres [65]. From grinding, RCF can achieve 1€/kg at a plant capacity of 1000 t/yr. However, higher recycling production capacity and higher carbon fibre recovery rate are required for advanced recycling technologies to compete with the virgin carbon fibres and RCF from more cost-effective recycling technologies available.

5 Recovered carbon fibre applications

5.1 Remanufacturing of recovered carbon fibres for structural polymer matrix composites

Approximately 40% of CFRP waste is generated during the manufacturing stage [66], out of which, dry carbon fibre wastes were repurposed into stitched non-wovens and manufactured into car seats and roof structures of BMW i3 and i8. The dry carbon fibres were also co-mingled with thermoplastic fibres to prepare either nonwoven or co-mingled yarns, for weaving into fabrics before hot pressing into thermoplastic matrix composites [67]. Chopped dry non-crimp fabrics have been recently processed into bulk

moulding compounds with comparable mechanical properties as existing SMCs that utilise virgin carbon fibres [68]. These reused carbon fibre materials will be particularly suited in automotive, construction, and aircraft interior applications.

The recovered fibres from CFRP waste are used in applications that require less stringent mechanical performance, albeit at a lower cost. **Fig. 8** shows a representative flow process of reusing recovered fibres from a high-cost aerospace composite component to a low-cost, non-structural component [23]. The closed-loop starts from an aerospace unidirectional FRP structural component. The recovered, chopped fibres are pressed to form an SMC automotive structural part. This part can be further recycled into a non-woven, short fibre SMC with lower fibre volume fraction to form a semi-structural component suited for automotive and marine applications. These short fibres can be further reclaimed and potentially used as a powder to reinforce thermoplastic matrices through injection moulding, which are not suited for structural applications in mass transit and automobiles. Today, remanufacturing methods are still emerging, which causes a deficiency in a discernible, viable end-use for the reclaimed RCFs [69].

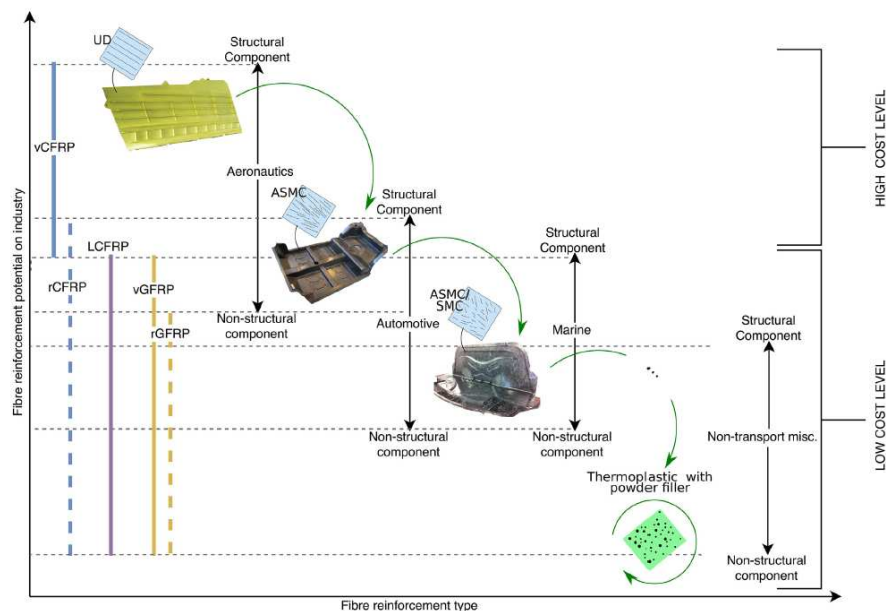


Fig. 8. A representation of reuse of recycled composite materials in closed-loop material flow at the low and high-cost levels with respect to their fibre reinforcement potential Reprinted from [23], Copyright (2019), with permission from Elsevier.

5.2 Mechanical performance enhancement of remanufactured polymer matrix composites

The recovered fibres from the CFRP end-of-life components have short lengths and hence low aspect ratios because of (a) the size-reduction of components before recycling, (b) the damage to fibres during recycling, and (c) the chopping process. The suitable processing technologies for the short fibre reinforcement are hence compression moulding and injection moulding, which are appropriate for the RCF particulate/powder. The greatest impediment in utilising RCF effectively is the lack of fibre alignment in the recycle. Optimization of fibre length, fibre orientation/alignment and the layer-wise relative stacking of discontinuous fibres are deemed as critical for achieving balanced mechanical performance in the remanufactured composite [70].

Fibre alignment is hence applied to increase the fibre volume fraction and to achieve improvements in the mechanical properties of the remanufactured composites. Explosives Research and Development Establishment (EDRE) in the United Kingdom devised and developed a patented process based on extrusion, filtration and centrifugal processes [71]. This patented process offers a more effective fibre alignment by using a centrifugal process that utilises a circular nozzle with improved filtration for quick removal of viscous liquid and reduced edge effect caused by the oscillation of a convergent head. Recently, the High-Performance Discontinuous Fibre (HiPerDiF) method [72, 73] and a patented I2M process [74] were developed to achieve alignment in discontinuous recovered fibres for the remanufacturing of composites with enhanced tensile properties in unidirectional [73] and quasi-isotropic laminates [75]. The HiPerDiF process works by subjecting the fibres in water to a sudden momentum change of the suspension, and therefore, aligning them transversely to a suspension jet. High fibre volume fractions of 55%, with 67% of fibres in the range of $\pm 3^\circ$ to the alignment direction were achieved. An average modulus of 71.8 GPa with 3 mm RCF length was realised [72]. The HiPerDiF method also allows the mixing of different types of fibres to realise pseudo-ductile behaviour, where fragmentation occurs progressively, but not catastrophically [75].

Oliveux et al. used carbon fibre tows recovered by solvolysis to align fibres and remanufacture composite laminates. The recovered fibre tows were aligned by a patented I2M process, which uses carbon fibre tows in a U-shape channel to produce a 25 mm wide tapes (**Fig. 9**). Their work showed that the RCF tows can compete with virgin fibres if good fibre surface and alignment are realised [74]. Nekoda et al. used a centrifugal alignment rig which allows control of solution temperature, drum rotation rate and nozzle linear velocity and achieved an increase of 100% and 137% in tensile strength and modulus over non-aligned composites, respectively [76].

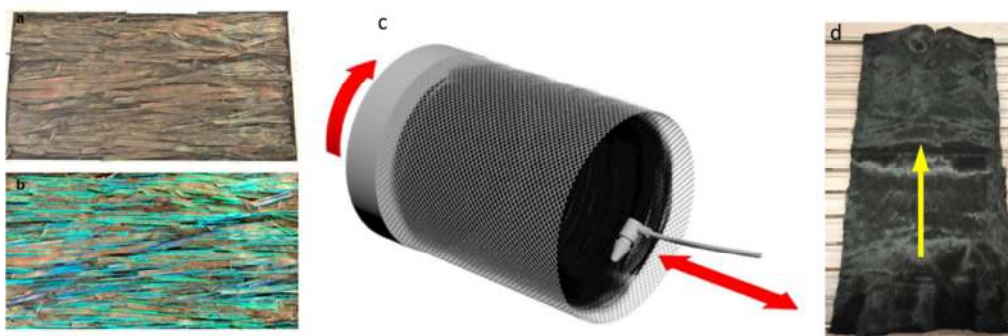


Fig. 9. Realigned recovered carbon fibre tows (a) as-obtained, and (b) fibres detected via green recolouration and recovered carbon fibre mat (c) collection cylinder and (d) aligned carbon fibre mat. Reprinted from [74] and [76], Copyright (2019 and 2020) with permission from Elsevier.

Reclamation and remanufacturing of virgin and recovered CF with polyamide 6 (PA6) was achieved by Tapper et al. [77] utilising the HiPerDiF alignment, as shown in **Fig. 10**. Compression-moulded discontinuous carbon (28 vol.%) in PA6 matrix was achieved with 39.7% reduction in tensile stiffness and 40.4% reduction in tensile strength upon remanufacture. Presence of fibre misalignment and agglomeration led to stiffness reduction in addition to incomplete fibre separation, and fibre breakage caused by high compaction pressures. The ultimate tensile strain was near-constant, indicating the polymer remained unaffected.

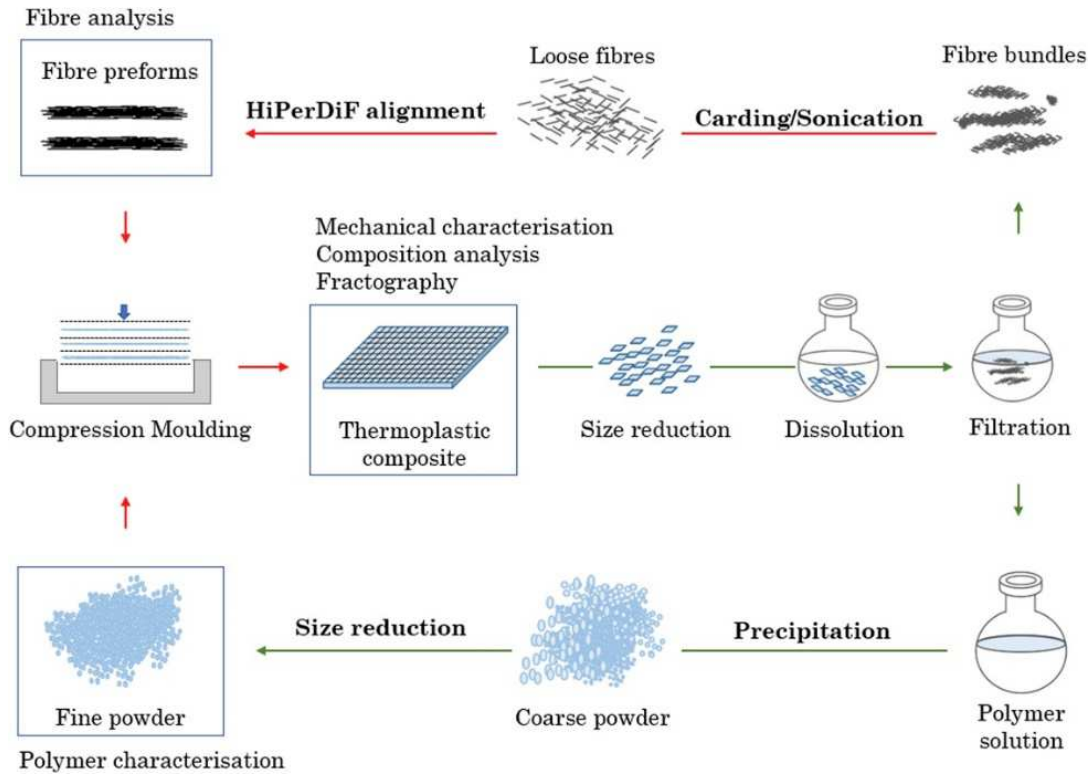


Fig. 10. CFRP recycling methods that utilise the HiPerDiF alignment technique, with green showing reclamation and red showing the remanufacturing flow. Reprinted from [61], Copyright (2019), with permission from Elsevier.

Fibre length distribution is also critical for remanufacturing of RCF based composites. Novel image processing and visualisation methods are being applied to the flake layout that is independent of the offcut geometry and orientation of feed into mechanical shredding equipment, and often is valuable if no prior layout information is available [78].

The mechanical performance of discontinuous CFRPs is often engineered through architecture and interlayer inclusions. Brittleness of discontinuous remanufactured CFRPs [70] is challenging because of the stress raisers in fibre ends. Strategies to alleviate this ductility include interlayer hybridization of discontinuous carbon fibres with self-reinforcing PP fibres, to compensate for the brittleness. In contrast to brittle failure, the use of self-reinforcing PP allows delayed tensile failure at $\epsilon_T \approx 10\%$. These interlayer hybrid CFRPs with self-reinforcing PP with a hierarchical, polygonal structure encompassing ‘cuts’ in the carbon fibre layer with concomitant, increasing

hierarchy caused a decelerated failure onset, thereby increasing the damage resistance in potential practical applications. Mechanistically, the fibre bundle fracture was achieved with spread out delamination along with well-spread fibre-bundle pull-out within the polygonal patterns. High stiffness and ductility were achieved through tailoring the pull-out behaviour of the fibre bundles and biomimetic design. Aligned, remanufactured CFRPs in the quasi-isotropic layup [75] revealed the improvements in tensile properties including the strain-to-failure, which were projected to bring about a higher level of improvements upon increasing the volume fraction of the RCFs to 55 vol. %. The HiPerDiF and other alignment procedures are central to these developments, and more novel alignment techniques are anticipated in the future adding to this critical, emerging field.

5.3 RCFs as reinforcement in cementitious mortars

RCFs reclaimed through mechanical recycling of thermoset CFRP can be reused in cementitious mortars, however, they often still possess residual thermoset polymer on the fibre surface, which decreases the interfacial adhesion to the matrix. Acid or alkali treatments are often more suitable for preparing carbon fibres for cementitious mortar reinforcement [79]. A chemical treatment especially is deemed to bring value addition to RCF composites through performance enhancements in the base cementitious matrix. Wang et al. [80] investigated the effect of a saturated simulated concrete pore epoxy RCF composites before use in cementitious mortar, with variable saturated simulated concrete pore solution, triisopropanolamine (1-[bis(2-hydroxypropyl)amino]propan-2-ol), and montmorillonite nanoclay, retaining the water-to-binder (w/b) ratio of all mortar mixtures at 0.50. The chemical treatment of RCFs showed enhancements in mechanical and chemical adhesion with the cementitious mortar. Wang et al. [79] produced RCF composites using wind turbine blade waste treated by NaOH into cement mortar. The inclusion of RCFs in mortars afforded higher strength and volume stability of the cementitious mortars. Mild NaOH (1 mol/L) provided a better adhesion effect over stronger concentrations, and untreated RCF mortars, in addition to reducing free drying shrinkage. Hence, RCFs can bring about necessary stability to the cementitious mortars, without affecting workability.

Conductive cementitious materials using RCFs were developed using four different types of PAN-based RCFs varying the aspect ratio in two concrete dosages of conventional and ultra-high-performance concrete mixes [81]. The use of RCFs was specifically promising for the ultra-high performance concrete mixes because the cost of the RCFs is significantly lower than comparable carbonaceous additives. Wet-mix method led to better workability and more uniform fibre dispersion. Electrical resistivity values in the range of 3–0.6 Ω m were obtained for 0.2 vol. % to 0.8 vol. % RCF inclusion, potentially positioning RCF as a candidate for multifunctional cementitious materials for industrial applications.

Carbon fibre/epoxy composites recycled without any pre-treatment can often result in increases in flexural strength to Portland cement, geopolymers, fly ash and metakaolin. Flexural strength increased without compromising compressive strength or additional porosity in a work by Saccani et al. [82], indicating the recycling being not critical for reuse or remanufacturing avenues. Prepreg scrap was used along with fly ash, metakaolin, and Portland cements up to 5 vol. % [82]. Workability of these composite admixtures decreased with increasing RCF fraction albeit with no compromise in the porosity. The moisture absorption decreased with an increase in the RCF fraction. Flexural strength and impact toughness increased, and the compressive strength was unaffected. The addition of RCF was deemed particularly critical for geopolymer matrices investigated.

Electrical conductivity and piezoresistivity were achieved through the addition of combined 0.2 vol % of graphene nanoplatelets to RCF in limestone-based mortars upon drying at the 28-day interval [83]. Optimising the current density and NaCl concentration allowed the mitigation of electrochemical corrosion and enhancement of RCF tensile strength. The use of nitric acid also served towards increasing the recycling efficiency and the interfacial shear strength, with higher temperatures identified to further intensify these effects.

Hence, the inclusion of RCFs into cementitious mortars causes increases in the mechanical stability and physical effects, while also providing a framework for creating multifunctionalities for potential industrial applications.

5.4 Non-structural applications

RCF substrates often provide a mechanically-stable framework for non-structural applications such as thermoelectric materials and flexible batteries. Jagadish et al. [84-86] used recovered Toray T600 carbon fibre obtained from fluidised bed process with multi-wall carbon nanotube (MWCNT) doping levels of 0.10 wt% and 0.15 wt% RCF-Bi₂Te₃ and RCF-Bi₂S₃ respectively [85] for a thermoelectric application. The MWCNT-doped RCF-Bi₂Te₃ and RCF-Bi₂S₃ exhibited power factors of 1.044 and 0.849 $\mu\text{W K}^{-2}\text{m}^{-1}$, respectively. The addition of MWCNTs enhanced the power factors of RCF-Bi₂Te₃ and RCF-Bi₂S₃ composites over 400% and 800%, respectively. Inclusion of MWCNTs allowed further improvements in the power factor (**Fig. 11**) of Bi₂S₃ thermoelectric composites comparable as Bi₂Te₃, with additional enhancements in crystallinity and thermal stability the RCF thermoelectric composites. N-type bismuth telluride (Bi₂Te₃) was electrodeposited as films on RCF, where the electrodeposition parameters of Bi₂Te₃ on RCF including deposition potential, deposition time, deposition temperature and electrolyte composition were determined as critical for the resultant Seebeck coefficient [84]. A multi-parameter interaction provided effective optimization in comparison to a univariate experimental design because of the presence of simultaneous effects of more than one deposition parameter.

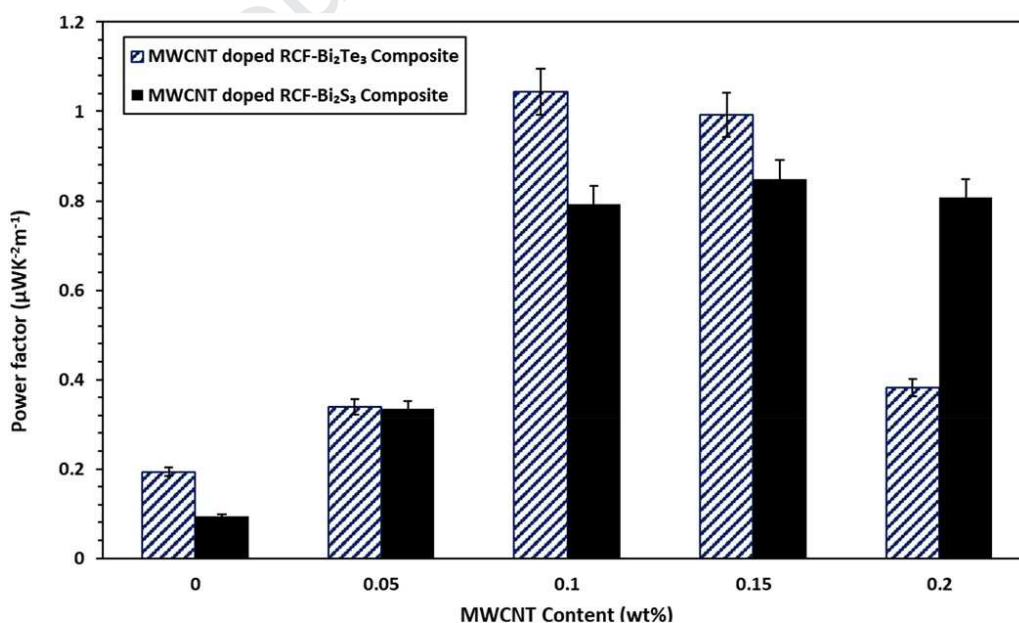


Fig. 11. A plot of the power factor as a function of MWCNT doping content in the RCF-Bi₂Te₃ and RCF-Bi₂S₃ composites. Reprinted from [86], Copyright (2019), with permission from Elsevier.

The widespread application of flexible lithium-ion batteries for wearables is still emerging [87], necessitating effective mechanical flexibility in addition to gravimetric energy density, rate capability, and cyclic life requirements. To this end, carbon spun-fabric electrodes (**Fig. 12**) were deemed suitable for high active material loading in the current collector that possessed intimate contact with the electrode by Cho et al. [88]. The recovered, oxidized PAN fibres were used to create a three-dimensional porous carbon spun fabric structure, which was scalable for use in garments. The structural, electrical, mechanical, and thermal analysis supported the use of these spun fabrics as heat insulation or heat sink. Moreover, the high surface area of the spun structure was conducive to LiFePO₄ loading, exhibiting improved stability and retention of rate capability over their aluminium counterparts. Hence, RCFs possess a strong potential to be used as a stable and flexible infrastructure for flexible lithium batteries.

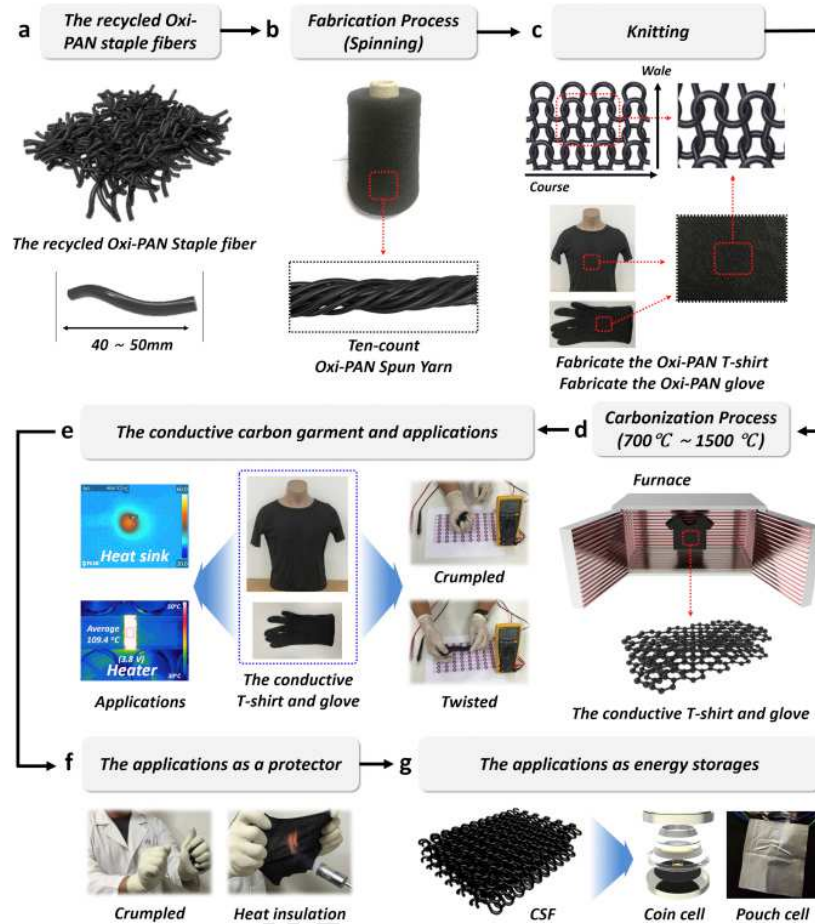


Fig. 12. A schematic of the flexible carbon spun fabric structures produced using RCFs. Reprinted from [88], Copyright (2019), with permission from Elsevier.

6 Development of recyclable matrix for CFRPs

The significant challenges associated with recycling CFRPs and the resulting environmental impact call for the development of recyclable materials. Research investigations on degradable thermosets and liquid moulding compatible, low viscosity thermoplastics are gaining much attention, which addresses the increasing concerns of sustainability and environmental responsibility of CFRP usage [89]. Natural fibres and biodegradable resins are also materials choice, however, they are mainly used for applications that do not require high mechanical performance and undergo severe weathering or harsh working conditions.

6.1 Degradable thermosets

Conventional thermoset polymers form stable cross-linking structure after the irreversible curing process and cannot be reshaped, reprocessed by heat, or decomposed under mild conditions. Removing the resin matrix from the carbon fibre or glass fibre reinforcement at the end-of-life of CFRPs is hence a challenging process. Lately, this challenge has received an immense response through synthesizing thermosets that can break down to simpler compounds or polymers to recover original materials. These degradable thermosets represent some innovative methods to radically solve composite recycling problems. In Ma and Webster's review of degradable thermosets based on labile bonds or linkages, the recent research was classified into polymers with different weak bonds or linkages: ester bonds, sulphur containing linkages, nitrogen-containing structures, orthoester structures, carbonate linkages, acetal linkages, hemiacetal/hemiketal ester linkages, olefinic bonds, Diels-Alder structures, vicinal tricarbonyl structures, peroxide bonds, phosphorous-containing structures, and tertiary ether bonds [90]. For example, the degradable thermoset based on tertiary ester bonds showed a glass transition temperature (T_g) of 73~185 °C, a tensile modulus of ~5.5 GPa and fracture toughness of 1.75 MPa m^{1/2} [91]. A polymer based on aminal bonds exhibited T_g of 125~200 °C, a tensile strength of 127 MPa, and fracture toughness of 1.94 MPa m^{1/2} [92]. The degradation stimuli for these degradable thermosets are temperature, UV radiation, and acidolysis. The weak stability of the bonds leads to the composites to exhibit inferior mechanical and thermomechanical properties than epoxy and bismaleimide (BMI) resins. Yuan et al. synthesized a new poly(hexahydrotriazine) (PHT) thermosetting resin and used it as a matrix for carbon fibre reinforcement [93]. The degradable thermoset showed a stable and strong covalent structure, which results in high mechanical performance and thermal stability as well as multiple recovery of carbon fibres and 88.6% reclamation of polymers through gentle depolymerization in a dilute acid solution.

Dynamic reversible bonds have been recently added to conventional thermosets such as epoxies to combine the high mechanical property and the characteristics of being able to be reshaped and reprocessed. Yu et al. realised near 100% recyclability of CFRP by synthesizing diglycidyl ether of bisphenol A (DGEBA) with fatty acids in the presence

of metal catalyst $\text{Zn}(\text{Ac})_2$, forming the epoxy covalent adaptive networks (CANs) that can undergo transesterification-cation-type bond exchange reactions (BERs) [94]. The epoxy CANs were shown to completely dissolve in ethylene glycol at a low temperature of 160~180°C. Further heating resulted in evaporation of solvents and repolymerization of the epoxy. The recovered carbon fibres maintained a high fraction of the mechanical properties, i.e., 97% of modulus and 95% of the tensile strength of the virgin fibres. Therefore, the reclaimed carbon fibres and recycled thermoset could be reused for preparing CFRPs. **Fig. 13** presents the schematic graph of the dissolution and repolymerization process.

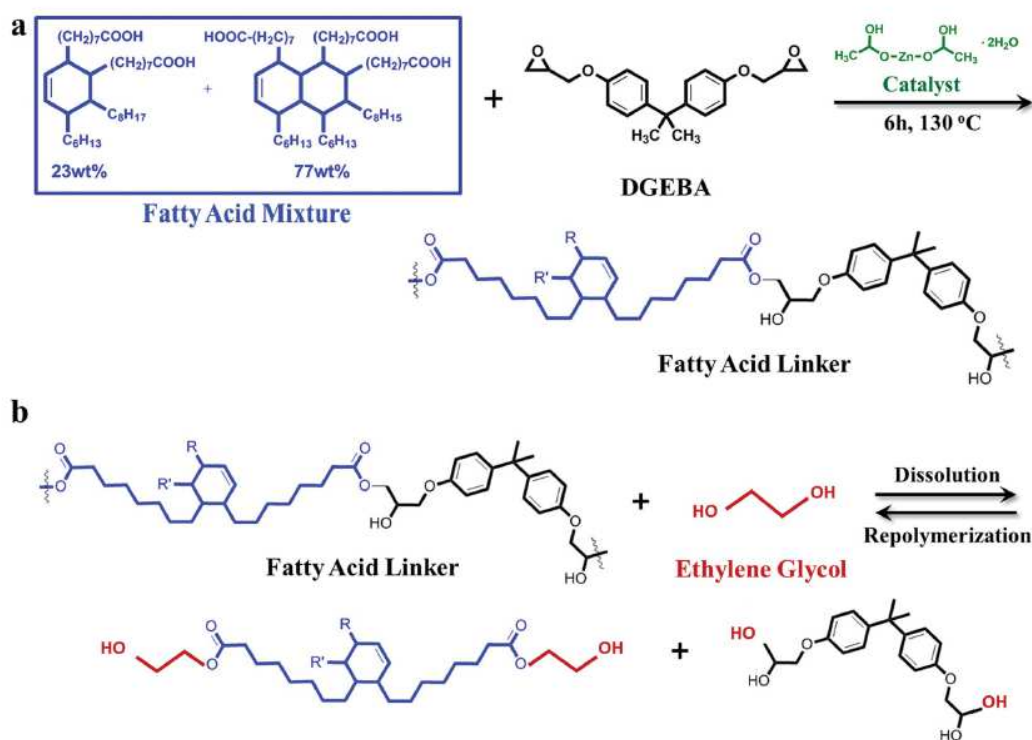


Fig. 13. Dissolution and repolymerization of the DGEBA epoxy with CANs in the ethylene glycol solvent, representing a) formation of fatty acid linkers within the thermosetting network, and b) dissolution and repolymerization of the epoxy CAN via transesterification. Reprinted from [94], Copyright (2019), with permission from Wiley.

Zhao and Abu-Omar reported recyclable and malleable epoxy thermoset bearing aromatic imine bonds without the need for metal catalyst or heating [95]. They attached imine bonds to the epoxy network to prepare the thermoset that has high

thermomechanical properties as well as being malleable. Through the breakage and reconnection of imine bonds, the epoxy was able to dissolve in organic solvents or slightly acidified water at 25°C and be reclaimed from solvents with comparable thermal and mechanical properties. At 80~120°C, the epoxy can be welded and repaired utilising the imine exchange reaction.

The availability of degradable thermoset matrices, along with RCFs are anticipated to bring about the essential sustainability characteristics for future CFRPs, which is an area of research that will gain prominence in the near future.

6.2 Thermoplastic matrix composites

The challenges of recycling thermoset matrix composites enable the high-volume usage of thermoplastics as the preferable option for lightweight components and structures. Although thermoplastic matrix composites are easy to be reshaped, reprocessed, and usually produced in short manufacturing cycles, they possess lower mechanical properties at the maximum operating temperature of most engineered structures, and with exceptions, are not compatible with low-cost liquid moulding technologies for generating complex component geometries. Low-viscosity thermoplastics with high mechanical performance are attractive for replacing thermosets to fabricate FRPs that are easily recyclable and bear a low environmental impact. Solvay company developed a novel polyamide 6,6 (PA66) with a viscosity of 15 Pa-s at melted state, suitable for resin transfer moulding (RTM) [96]. This PA66 exhibits improved mechanical properties, T_g , and lower water absorption due to modification with phenol-formaldehyde. Rottger et al. used a metathesis of dioxaborolanes to prepare crosslinked vitrimers from different thermoplastics such as polystyrene and high-density polyethylene, which are processable via extrusion and injection moulding for multiple times [97]. These resins show superior chemical resistance and dimensional stability and possess the potential to attain high mechanical and thermal performances. A rapid and thermally robust synthesis strategy is suitable for polymers with backbones made of carbon-carbon single bonds. The potential availability of the chemically- and thermally-resistant thermoplastics will bring about a transformation in the design methodology of applications that utilise thermoset matrices traditionally.

In thermoplastic composites, a divergent range of sizings is utilised to prepare virgin CFs or RCFs for use with thermoplastic matrices, especially for carbon fibres with poor wettability and adhesion to thermoplastic matrices. A review of different surface treatment methods of carbon fibres is presented in [98] for reinforcing thermoplastic matrices including polyethylene (PE), polypropylene (PP), and acrylonitrile butadiene styrene (ABS) resins, polyamide (PA), polycarbonate (PC), polyetheretherketone (PEEK), and polyetherimide (PEI).

Often, RCF use in thermoplastic FRPs is offset or aided by the use of other functional phases, for example, biobased pyrolysed carbon used in 5 – 20 wt. % to enhance the mechanical properties of PC resin [99]. An RCF/biocarbon ratio of 1:1 led to increases in mechanical properties including impact strength. RCFs were also applied to wear-resistant poly(ether ketone) composites processed using co-rotating twin-screw extrusion [100]. While the RCF composite showed marginally lower mechanical properties, RCFs were particularly effectual reinforcements that cause enhanced friction and wear properties under moderate and severe pressure and surface velocity conditions. The authors indicated that the RCFs can replace VCFs without a major loss in wear performance, hence creating opportunities for RCFs in tribological applications. Hence, the rationale of RCF use in thermoplastic FRPs is well supported in static, impact, and tribological performances.

7 Conclusion

Composite recycling is a critical issue for the sustainable use of lightweight composite materials in the fields of transportation, construction, and sports equipment. Current recycling methods include mechanical recycling, high voltage fragmentation, pyrolysis, fluidised bed, chemical/thermochemical recycling by solvolysis. The alignment of reclaimed long fibres is shown to enhance the mechanical performance of the remanufactured CFRP significantly. New and economical recycling technologies are urgently needed to effectively reclaim valuable carbon fibres and glass fibres. In this regard, several promising techniques have been demonstrated as better and more economical recycling solutions, which utilise the inherent energy within composites to decompose thermoset matrices, use mild treatment conditions involving less

environmental damage, and retain reinforcement properties of fibre, maximise the performance of remanufactured composites from reclaimed fibres with/without recycled polymer matrices. Materials research in developing new degradable thermosets and liquid moulding compatible, low viscosity thermoplastics also hold tremendous promises for a future circular economy. More radical approaches such as replacing thermoset matrix with new thermoplastics are feasible for composites in the wind turbine and other relevant sectors.

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Declaration of interests

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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