Resources, Conservation & Recycling 166 (2021) 105340

Contents lists available at ScienceDirect

Resources, Conservation & Recycling

journal homepage: www.elsevier.com/locate/resconrec

Review

Recent progress in recycling carbon fibre reinforced composites and dry carbon fibre wastes

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ARTICLE INFO

Keywords:
Carbon fibre
Recycling CFRC
Life cycle assessment (LCA) analysis
Dry carbon fibre waste
Carbon fibre yarns and nonwovens

ABSTRACT

Carbon fibre reinforced composites (CFRC) continue to play a key role in the growth and development of many weight sensitive industries. However, their proliferation has raised increasing concerns regarding the required practices and strategies to deal with these expensive engineered structures at the end of their life cycle. This has brought into focus the need to develop more sustainable and efficient recycling solutions for these products. Similarly, the necessity of managing dry carbon fibre scraps generated during the manufacturing process of CFRC has attracted more attention in recent years. This review article provides an overview on recent advances in recycling CFRC as well as processing dry carbon fibre scraps. Influential parameters, advantages, drawbacks, and possible environmental impacts of the main technologies of processing CFRC waste including mechanical, thermal (pyrolysis and fluidised bed), and chemical (solvolysis and low temperature chemical processing) will be evaluated. Their potential effects on mechanical characteristics and surface chemistry of fibres are assessed. Moreover, recent processing methods of dry and semi-finished carbon fibre scrap are also reviewed. Specific attention is paid to the recent developments in producing hybrid yarns and nonwovens made of waste carbon fibre. The methodologies developed in this area, their processing conditions, as well as other important findings are discussed. This review paper provides a valuable platform for researchers and decision makers working in the field of carbon fibre by providing a clearer picture on the options available to recycle CFRC, and the methods of developing value-added products using waste carbon fibre.

1. Introduction

The development of carbon fibre-reinforced composites (CFRC) has attracted significant attention in recent decades primarily because of their lightweight and superior mechanical properties. These engineered fibres and resultant composites have been employed in various fields such as aerospace, automotive, construction, and energy where their properties are most advantageous (Cousins et al., 2019; Hadigheh et al. 2018). CFRC are materials in which the embedded fibres act as load-carrying elements while the polymeric matrix stabilises their orientation, and protects them against environmental damage (Lau et al., 2018; Yao et al., 2018). Carbon fibres (CF) have played a key role due to their superior strength to weight ratio making these composites lighter, stronger, and more durable. The mechanical specifications of CF depend on the precursor type and production procedure which namely are polyacrylonitrile (PAN) and coal pitch carbonisation. These fibres are used in different forms of long or short filaments, chopped, milled, woven, and nonwoven mats, for manufacturing CFRC (Alam et al., 2019). This has led to many applications by replacing conventional materials such as steel, aluminium, and alloys in different industries (Yao et al., 2018; Verma et al., 2018; Clark et al., 2020). Specific additional advantages of carbon fibre composites include its durability in aggressive conditions and at high temperatures, abrasion resistance, and corrosion resistance (Clark et al., 2020; Francis et al., 2017; Hadigheh et al. 2020).

The demand for CF has continued to grow, at a current annual growth rate of around 11% due to the increased need for lightweight materials, and it has been estimated that total global demand for CF could reach up to 117,000 tonnes in 2022 (Lin and Schlarb, 2019; Zhang et al., 2020; He et al., 2020). Such a high demand however, introduces disadvantages, and will dramatically increase the amount of CFRC waste in the coming years, raising concerns regarding the management of these composite products at the end of their service life (Lin and Schlarb, 2019; Naqvi et al., 2018). In the wind turbine sector for

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https://doi.org/10.1016/j.resconrec.2020.105340
Received 18 November 2020; Accepted 4 December 2020

instance, it is estimated that the cumulative waste of CFRC utilised in manufacturing process of turbine blades will reach 483,000 tons by 2050 in Europe alone (Lefeuvre et al., 2019). The use of CF in this area is increasing, as it enables the design and manufacture of larger, more efficient, lighter and stronger blades with higher power generation capacity compared with those prepared with glass fibres (Lefeuvre et al., 2019). Similarly, there is a huge worldwide demand to use CF in other industries such as the automotive where the total value of CF was approximately USD 2.4 billion in 2015 and expected to increase to USD 6.3 billion by 2021 (Meng et al., 2018). Based on the European directive 2000/53/EC, from 2015 onward, it is required to recover and reuse 95% of the total vehicles components and recycle at least 85% of them at the end of their life cycle (European Council 2000; La Rosa et al., 2016). Similar concerns have also been raised for composite parts in the aerospace industry where around 6000–8000 aircrafts will reach their end of service life by 2030, generating a significant amount of composite waste (Meng et al., 2018; Khalil, 2018; Longana et al., 2016; Vieira et al., 2017).

A major disadvantage of CF is that the production of virgin carbon fibre (vCF) is an expensive and energy intensive process which has negative ramifications on the environment and human health through the emission of hazardous by-products (Grzanka, 2014; van de Werken et al., 2019). The high-energy requirement of CF production comes primarily from the oxidation of the precursor prior to carbonisation (Morin et al., 2012). Life cycle assessment (LCA) analysis has shown that the embodied energy of CF, that amount of energy required for manufacture, is very high compared with other structural materials (Tapper et al., 2020). 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 (Song et al., 2009). Furthermore, the production of vCF creates gases such as hydrogen cyanide (HCN), ammonia (NH_3), carbon monoxide (CO), carbon dioxide (CO_2), nitrogen oxide (NO_x) and volatile organic compounds (VOCs) which need to be managed during the production process (Grzanka, 2014). In contrast, the embodied energy of recycled carbon fibre (rCF) is much less than that of the manufacturing process of vCF, although this will vary depending upon the recycling method. Clearly then, the embodied energy along with economic and environmental impacts, are all factors which are driving research into developing new and improved recycling solutions and applications for CF and CFRC. It is also estimated that by 2022, there will be a shortfall in supplying vCF of around 24,000 tonnes worldwide, which may be covered by rCF provided that the challenges of current recycling technologies can be overcome (Francis, 2019).

This article provides new outlooks to CFRC recycling and dry CF waste processing by shedding light on some important aspects on these areas. Firstly, it provides a comprehensive and comparative review on recent advances and modifications of different CFRC recycling methods, and discusses the impacts of each recycling route and associated parameters on recovered fibres chemistry and mechanical characteristics. Secondly, a specific discussion has been provided to compare economic and environmental benefits of different recycling methods in the context of LCA, which can provide a more general insight into advantages and disadvantages of each method. Finally, this paper provides an in-depth review on recycling dry carbon fibres waste which is an important part in carbon fibre waste processing chain. Very little attention has previously been paid to processing dry carbon fibre wastes and producing value-added products. It is estimated that the dry CF scraps account for around 40% of the total waste in this area (Pimenta and Pinho, 2011), showing the importance of its processing for further flourishing of carbon fibre recycling industry. A critical evaluation of the methodologies, research gaps and recommendations for future research will also be presented in this article.

2. Recovery methods of CFs from CFRC

In general, an appropriate recycling process should ideally be efficient, eco-friendly, cost-effective, and with minimum impact on fibres length and interfacial compatibility with new resins (G. Oliveux et al., 2015). There are three main types of CF waste which include i) carbon fibre offcuts generated from production (dry fibres) whose mechanical characteristics are similar to vCF, ii) prepreg residues and semi-finished products, and iii) fibres which can be reclaimed from CFRCs (Saburow et al., 2017; Pickering, 2006). In respect to CFRCs, there are three main strategies to manage the waste; disposal (landfilling), energy recovery (incineration) and recycling (e.g. mechanical, chemical and thermal methods) (Khalil, 2018; Rybicka et al., 2016). The landfilling is probably the most common and cheapest method of disposal; however, this is not considered as an eco-friendly approach and both US and EU authorities have put restrictive legislations in place on landfilling CFRCs (Khalil, 2018; Tapper et al., 2018). Indeed, it is not possible to recover the embodied energy of the composite materials through this method. Incineration can be used to recover some energy from the waste products; however, it releases large amounts of pollutants into the environment which is not considered to be a sustainable solution for managing the waste in the long term (Gharde and Kandasubramanian, 2019). Other approaches have therefore been developed to minimise the impact of CF waste on the environment and to create new high value-added products.

The recycling of CF has largely consisted of fibre recovery from the polymeric matrix, then remanufacture of the recycled fibres into a useful product (Pickering, 2006; Y. Liu et al., 2017). The recycling of composites depends in large measure on their polymer type and whether the matrix is thermoplastic or thermoset. The former group, in principle, can be re-melted, re-shaped and re-moulded to manufacture new products (Y. Liu et al., 2017). However, for the composites made of thermoset matrix, the recycling process is more complex due to their heterogeneous nature and difficulty in liberating the fibres (Pickering, 2006; Y. Liu et al., 2017). Thermoset matrices such as phenolic resins or epoxy resins cannot be re-processed, recycled, or altered into their monomeric state due to their 3D cross-linked structure (Wang et al., 2018). The new processes developed to liberate the embedded CF from a thermoset matrix generally focus upon depolymerisation of the matrix into complex mixtures of gases, liquids and solid chemicals (Wang et al., 2018).

There are three main CF recovery approaches including thermal, chemical, and mechanical (Khalil, 2018; G. Oliveux et al., 2015). The thermal method includes pyrolysis, and fluidised bed pyrolysis. Some methods such as solvolysis, acid digestion, and supercritical fluid solvolysis, are amongst the main chemical approaches for liberating carbon fibre from composite materials. The mechanical method is mainly based on grinding the CFRCs into small pieces through shredding, grinding or milling. Although these recycling methods can be promising in terms of lowering the emission of greenhouse gasses, energy usage, and resources depletion, their financial viability is directly related to the cost of the consumed energy and the value of the obtained materials. Employing efficient recycling and re-use processes for CF can effectively close the loop of production and consumption of CF in different areas and can play a significant role in reducing the accumulation of CF waste. In addition to these CFRC recycling methods, there are some other methods such as high voltage fragmentation, electrohydraulic fragmentation, and electrically driven heterocatalytic decomposition (Leißner et al., 2018; Orzol et al., 2017; Zhu et al., 2019). Fig. 1 shows the main strategies for managing CFRC waste and dry CF scrap. Each of these waste management methods leads to different fibre length ranges, surface characteristics, and environmental impacts, all of which will be reviewed in this paper.

2.1. Mechanical recycling method

The mechanical processing of composites is a practical technique to

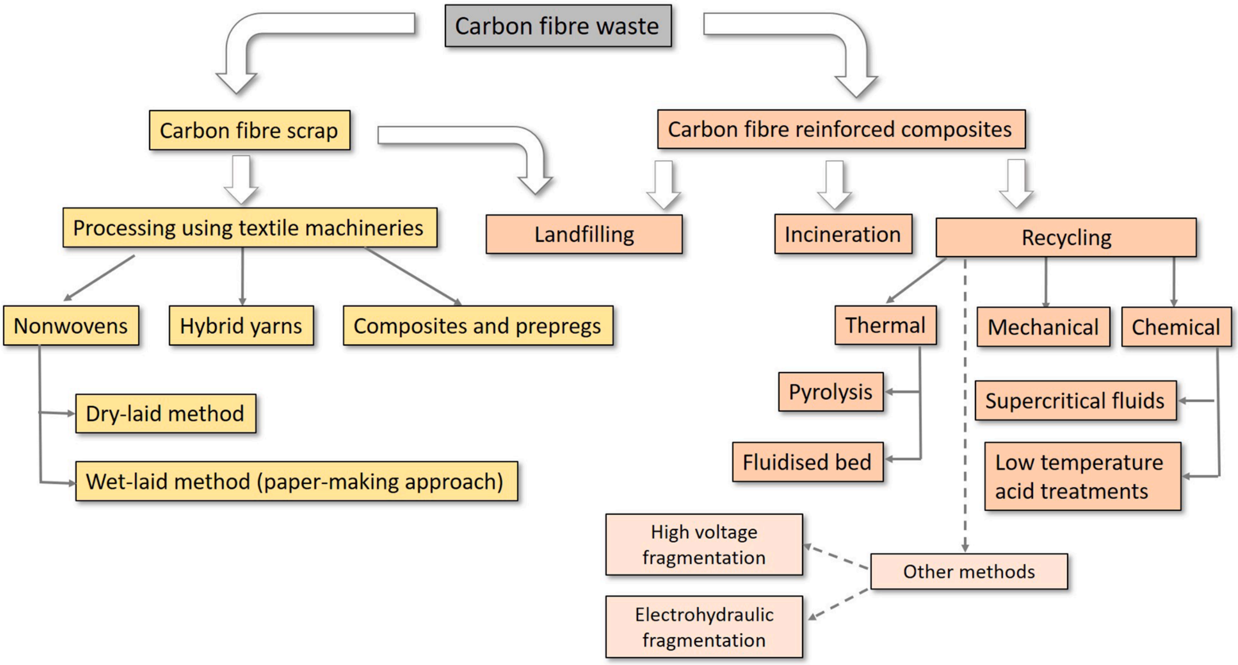

Fig. 1. CFRC waste and dry CF scrap management routes.

manage the increasingly large volumes of carbon fibre waste. This method is mainly based on shredding, crushing, grinding, and milling of the composite part into smaller pieces which can then be further ground into powder. To do this, equipment such as multiple shaft shredding and cutting mills have been used (Fig. 2). The shaft shredding produces large and uniform pieces controlled by the distance between the blades, the sieve size beneath the blades and their rotation speeds (Vincent et al., 2019) After producing flakes of composites and removing any metal parts, the shredded parts can then be ground into powder which is then remanufactured into a composite. For instance, flakes obtained from shredding a thermoplastic composite, CF-reinforced PPS (polyphenylene sulphide) has been re-used after blending with new fillers in a low shear mixer followed by re-shaping using compression moulding (Vincent et al., 2019). The low shear mixing was carried out under controlled heating to prepare a dough-like material, and to avoid breakage of fibres contributing to the retention of the mechanical properties of the remanufactured parts (Vincent et al., 2019; De Bruijn et al., 2017).

The mechanical recyclability and effects upon mechanical properties

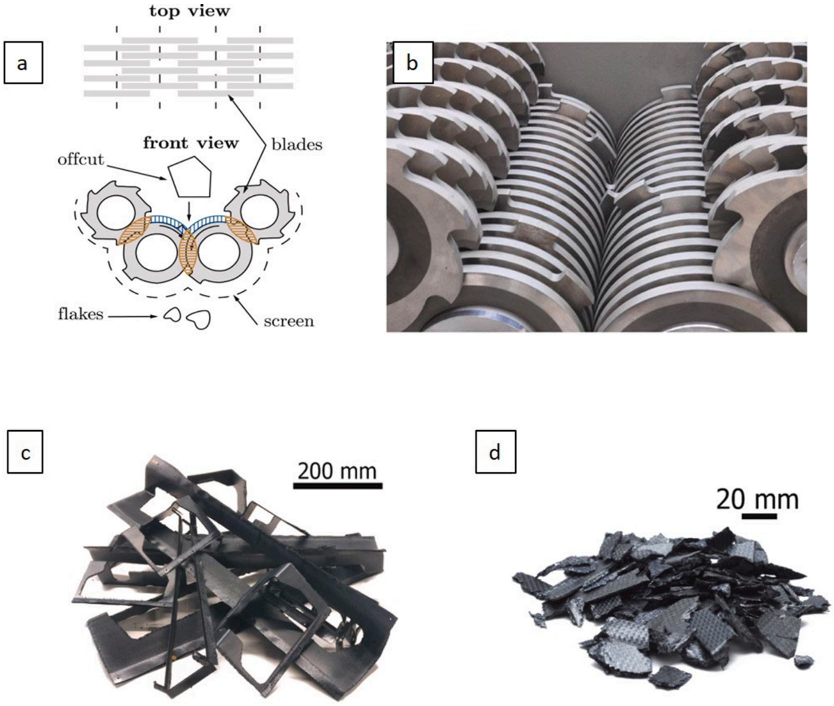

Fig. 2. a) Schematic illustration of top and front, and b) inside views of a multiple-shaft shredding machine; CF/PPS offcuts c) before and d) after shredding (Reprinted with permission from ref (Vincent et al., 2019), Copyright 2017 Elsevier).

of thermoplastic composites made of polyamide 66 (PA66) filled with short CF were investigated by Colucci et al. (Colucci et al., 2015). They discussed the effects of CF length shortening caused by recycling process along with UV-induced photodegradation on deteriorating the mechanical properties of the remanufactured parts. Initially, the composites were made in the form of bone-like tensile test bars by melting PA66 granules containing 30 wt% of 0.3 mm CF. Then, they were exposed to an accelerated ageing process under UV with varying temperatures and moisture to simulate the real ageing process (Colucci et al., 2015). The aged composites then underwent a recycling process including re-granulation, re-melting and re-injection moudling to make new recycled composite parts (Fig. 3). This artificial ageing process resulted in a reduction in elastic modulus and tensile strength and had a greater impact compared with the mechanical recycling process (Colucci et al., 2015). The reduction in mechanical properties arose from photo-oxidative and thermo-oxidative chain scission reactions in the polymeric chains caused by UV irradiation. After recycling, the overall CF length in the produced parts was shorter; however, this had a negligible impact on the mechanical properties of the recycled composites (Colucci et al., 2015). In addition to these findings, the role of some other factors such as fibre dispersion in the matrix, the orientation, and the optimised injection moudling procedure should be examined.

For effective mechanical processing of thermosetting polymer composites, the size of waste fragments should be reduced to less than 100 mm using low speed crushing mills. The metal parts and other impurities can be separated at this stage. A high speed hammer mill will then further downsize the fragments using impact and shearing actions (Francis et al., 2017; Shuaib and Mativenga, 2016). It has been demonstrated that the quality of recyclates and the consumed energy are affected by the clearances between the machine blades, the thickness of the fed composites and the sieves mesh size (Shuaib and Mativenga, 2016). The outcome of this process will be a mixture of fine fibres with the lengths of about 2–20 mm, powders and coarse recyclates containing polymeric particles with the approximate size of <100 μm. These can be separated via the sieving process into different sizes, obtaining resin-rich and fibre-rich pieces. However, there are limited applications for re-use and currently there is no developed market for these products. One of the potential applications could be to introduce them as fillers into composites, concrete, asphalt or coatings which is generally considered as an uneconomical down cycling (Pickering, 2006). Although there are some advantages for mechanical recycling method such as fast processing and facile scalability, it dramatically breaks the CF's length and mainly produces fibres which contain resin residues on their surface, which in turn lowers their reusability in new CFRC products. Producing recycled fibres with non-uniform lengths and surface characteristics is also another challenge associated with using this method.

There are few studies published in this area to evaluate the operating parameters and developing new applications. More attention however has been paid in literature to the mechanical recycling of GF reinforced composites rather than the CF-reinforced parts (Pietroluongo et al., 2020). In one of the early studies, Palmer et al. (Palmer et al., 2010), investigated producing value added products by using rCF powders as fillers in sheet moulding compound (SMC) systems to displace glass fibre. The powders were produced using a TRIA rotating hammer mill followed by separation into four different grades (Fig. 4). The obtained fine carbon fibres (part C) were used instead of GF in fabricating the SMC composites. The mechanical properties of the composites made using rCF was found to have a similar mechanical performance at 20% by volume of GF (Palmer et al., 2010). Similarly, Thomas et al. (Thomas et al., 2014) reported that adding 20% powder containing CF with the length of 1.25 mm, collected from the cutting of composite laminates, to resin increased compressive strength, flexural strength and modulus of elasticity by 20%, 30% and 30%, respectively. More recently, CFRC recyclates obtained from mechanical processing were re-used as reinforcing ingredients in the epoxy foams where they showed a very high impact. Adding 10wt% of recyclates, without segregation of fibre-rich and resin-rich components, to the epoxy foam led to 98% and 75% improvement in compressive modulus and strength, respectively (Song et al., 2020). The economic and environmental impacts of mechanical recycling will be discussed in Section 3.

2.2. *Thermal recycling of carbon fibres*

In this method, embedded fibres are released from the cross-linked polymeric matrices through thermal treatment which degrades the polymer matrix. The two main approaches to reclaim the CF are pyrolysis and fluidised bed methods. The following sections will discuss each of these methods, their impacts on surface and mechanical characteristics of fibres and recent developments in each area.

2.2.1. *Pyrolysis*

Amongst different methods of recycling of composites, pyrolysis has progressed to industrial level in recent years. In general, the pyrolysis process refers to the breaking down of polymeric matrix in an inert

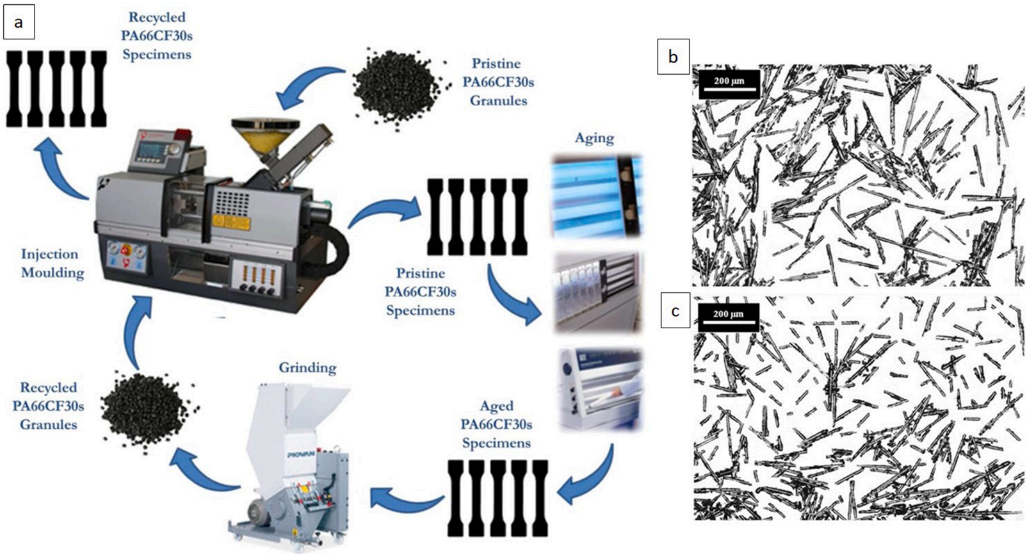

Fig. 3. a) the production procedures of PA66/CF composites and their mechanical recycling; optical images of CF b) before and c) after recycling process (Reprinted with permission from ref (Colucci et al., 2015), Copyright 2015 Wiley Periodicals, Inc.).

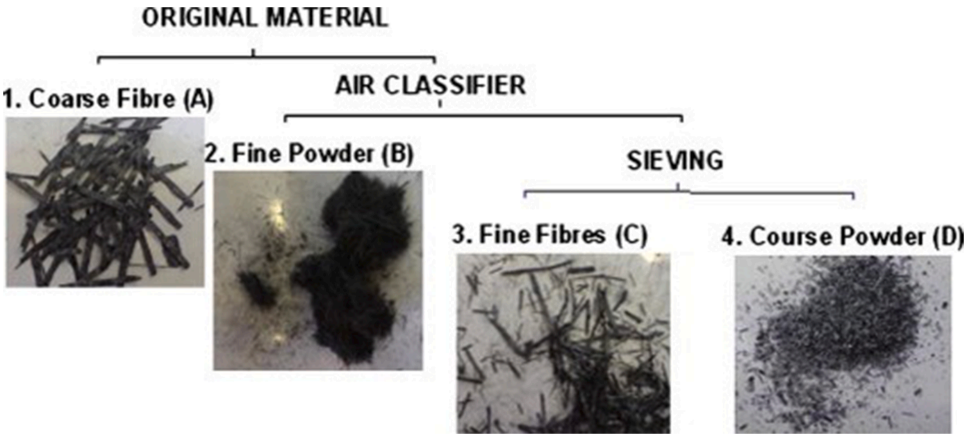

Fig. 4. Carbon fibre recyclates in four grades produced by mechanical processing (Reprinted with permission from ref (Palmer et al., 2010), Copyright 2010 Elsevier).

atmosphere, at a controlled temperature ($>$ 350 °C) and generally at atmospheric pressure (G. Oliveux et al., 2015; Meyer et al., 2009). In pyrolysis, the matrix is decomposed into the mixture of solids (fibres and fillers), oil (e.g. benzene, toluene, ethyl benzene and phenols), gas (e.g. H_2, CH_4, CO and CO_2) and polyaromatic chars (Naqvi et al., 2018). The generated liquids and gases have various molecular weights and due to their high calorific values, can be collected and used as fuel or secondary resources for different manufacturing steps (López et al., 2013). Pyrolysis is a viable approach capable of producing recycled carbon fibre on a large scale. It is estimated that the total energy consumption in this method is around 10–50% of the total energy required for the production of vCF (Witik et al., 2013). In addition to pyrolysis, post pyrolysis oxidation is necessary to eliminate the pyrolytic char layer formed on the fibre surface after pyrolysis of the polymer (Naqvi et al., 2018). This post-gasification can cause negative effects on mechanical characteristics of the fibre, resulting in reduced tensile strength and elasticity (López et al., 2013; Onwudili et al., 2016; Yang et al., 2015). This can be minimised however, through optimisation of the operating conditions such as pyrolysis and oxidation temperatures, residence time and reaction atmosphere components. It has been reported however that the resultant composites made by rCF tend to have inferior overall mechanical properties compared with vCF-based composites. In addition, other characteristics of the recovered fibres through pyrolysis are not as consistent as vCF. For instance, some features such as crystallinity, sizing, and impurities may vary in each of the recovered fibres. In thick and multilayer feedstock cases, it has been reported that the characteristics of fibres can differ in outer and inner parts of the processed CFRC, where the latter one still contains some resin residues (Mazzocchetti et al., 2018). Therefore, the rCF obtained through this method has not been suggested as a perfect substitute for vCF with the composites only being suitable for non-structural applications such as internal panels in automotive industry and lightweight sports equipment (Onwudili et al., 2016; Mazzocchetti et al., 2018).

As mentioned, the pyrolysis recycling method consists of thermal degradation of the matrix and post-oxidation which have different impacts on surface chemistry and mechanical characteristics of fibres. One of the most important parameters which determines the quality of the recovered fibres is the temperature used to pyrolyse and oxidise the fibre. The possible impacts of these factors on mechanical characteristics of carbon fibres have been investigated by Nahil and Williams (Nahil and Williams, 2011). The composites made of woven carbon fibres (55–60%) and polybenzoxazine resin (40–45%) were pyrolysed at 350, 400, 450, 500 and 700 °C for 1 h in a static bed reactor (Nahil and Williams, 2011). Then, the post-oxidation process was carried out at 500 and 700 °C to eliminate any formed char layer on fibres. The results showed that the best retention of mechanical properties was at a pyrolysis and oxidation temperature of 500 °C, where 93% and 96% of the tensile strength and Young's modulus were maintained. By increasing the oxidation temperature, the required time to eliminate the char layer from fibre surfaces was shortened. For instance, the required time for the elimination of pyrolytic chars from the fibres reduced to 15 min from 2 h after increasing the oxidation temperature to 700 °C. However, it further deteriorated the mechanical characteristics of fibres where the best retention of tensile strength was only 36%. In addition, oxidation time is effective in determining the level of damage imposed on fibres and their final surface oxygen content (Mazzocchetti et al., 2018). This effect was studied by Mazzocchetti et al. (Mazzocchetti et al., 2018), where they monitored the effect of oxidation on both vCF and pyrolysed fibres. The results showed that the vCF was actually more prone to damage during the oxidation process compared with pyrolysed fibres. This was due to the presence of a char layer on fibres which resulted in 10% larger fibre diameter compared with vCF. Analysing the effect of oxidation process on vCF and pyrolysed fibres revealed that the char layer actually played a protective role on fibres, reducing the overall damage to the fibres (Mazzocchetti et al., 2018). The char acted as a sacrificial layer and prevented the reaction of oxygen with carbon atoms available in fibre structure, lowering the degradation pace of fibres through conversion of C atoms to CO and CO_2 (Mazzocchetti et al., 2018). The vCF initially showed 2.8% diameter reduction after undergoing oxidation at 500 °C for 20 min, which was due to the removal of sizing layer. After extending the oxidation time to 60 min, fibres underwent 10% diameter reduction due to the fibre degradation. This is while the oxidation of pyrolysed fibres for 20 min was not even effective to remove the char layer necessitating an extension of oxidation time to 60 min (Mazzocchetti et al., 2018). Analysing the fibre diameter after this oxidation treatment showed that the pyrolysed fibres had comparable diameters with vCF after the heat treatment at 500 °C for 20 min. These findings demonstrated the protective role of char layer on pyrolysed fibres against an excessive degradation process. Therefore, optimising the pyrolysis and oxidation parameters is required to maximise the retention of fibre mechanical features during recycling process (Mazzocchetti et al., 2018).

The impact of pyrolysis on fibres surface oxygen content has also been taken into account. The generally held view in literature is that the oxidation process can enrich the pyrolysed fibres surface with oxidised groups which can act as cross-linkers for rCF in contact with new resin. Therefore, in some cases even similar mechanical performances in terms of Young modulus, strength and elongation were reported for epoxy composites containing vCF and rCF (Mazzocchetti et al., 2018). Jiang and Pickering (Jiang and Pickering, 2016) also corroborated the role of oxidation process in introducing new oxidised groups; however, they discovered that the combined effects of pyrolysis and oxidation processes reduced overall oxygen content on fibres. They reported that O/C ratios of vCF and rCF based on XPS analysis were 0.258 and 0.15, respectively. During pyrolysis process, carbon fibres' surface oxygen functionalities were removed in the form of CO and CO_2. This loss would be compensated by the oxidation process; nevertheless, the overall oxygen content introduced at this step was less than that of the original fibres. This could contribute to lowering the interfacial shearing strength of recycled fibres after contacting with the new resin (Jiang and Pickering, 2016).

2.2.1.1. Some modifications in pyrolysis process. Effect of oxygen: Researchers have attempted to improve the efficiency of pyrolysis recycling method by implementing modifications such as using controlled concentration of oxygen gas, superheated steam, and microwave irradiation in the conventional pyrolysis recovery process. For instance, Yang et al. (Yang et al., 2015) studied the role of oxygen concentration, reaction time and temperature on surface chemistry and mechanical characteristics of rCFs. The results demonstrated that a delicate balance between these factors is required to recycle fibres without deteriorating their intrinsic characteristics (Yang et al., 2015). In general, the temperature of the process and the reaction time should both be adjusted based on the ratios of the gases introduced to the reactors. The pyrolysis process of composite made of 4,4-diaminodiphenylmethane cured epoxy resin was carried out at different gas mixtures including 100% N_2, 5% O_2–95% N_2, 10% O_2–90% N_2, and 100% air with the velocity of 600 ml/min at 550, 600 and 650 °C for 15–60 min. It was realised that by increasing the concentration of oxygen, temperature, and pyrolysis duration, a higher CF mass loss took place which was due to higher removal of pyrolytic carbon char layer as well as the oxidation of CFs (Yang et al., 2015). However, the dominant factor in causing the mentioned mass loss of fibres varied depending on the temperature and available oxygen. The temperature showed the key effect in low concentrations of oxygen, while its effect in air was negligible. Analysing the impact of oxygen content on tensile strength and tensile module of rCF revealed that increasing the oxygen gas concentration from 5% to 10%, decreased tensile strength rapidly, but further increasing to 20%, caused the reduction to level off, despite the tensile modulus starting to decline (Yang et al., 2015). Thermal processing at 650 °C in the presence of 5% oxygen for 45 min therefore was considered to be the optimum condition where the rCF showed 80% retention in tensile strength (Yang et al., 2015). Analysing the chemical composition of rCF showed that the concentration of oxygen gas was the dominant factor in determining the oxygen content on fibres surface, where increasing the oxygen amount in reactor led to higher O/C levels (Yang et al., 2015).

Superheated steam: There are some research studies on using superheated steam in the pyrolysis process of CFRC (Wada et al., 2016; Kim et al., 2017; Jeong et al., 2019). Kim et al. (Kim et al., 2017) modified the pyrolysis process by exposing the CFRCs to superheated water steam at 550 °C for 30 min. This was followed by an oxidation process in air at 550 °C for 30–75 min. In this method, instead of forming volatile compounds, high molecular weight tar was produced which therefore contributed to less emission of carbon dioxide. A clean surface of rCF was achieved after 75 min of oxidation in air, while shorter oxidation periods led to remaining resin residues on the fibre surface. The tensile strength of fibres dropped by 10–15% after only 30 min processing and further extending the time to 60 min did not change this trend significantly. This method led to the retention of around 90% of the fibre tensile strength after 60 min treatment. In terms of surface elemental composition, steam led to a higher oxygen content on the surface through the introduction of –OH, –CH–, H–bond, $-C=O$ and C–O functional groups, resulting in a more polar surface with higher surface energy (Kim et al., 2017). This was attributed to the improved adhesion capability of fibres when manufactured with fresh resin. Measuring the interfacial shear strength (IFSS) of composites made of rCF demonstrated a higher IFSS level compared with vCF-based one. The overall oxygen content was also affected by the heat treatment time and prolonging it slightly lowered the IFSS level for the single rCF composites. The initial increase was referred to a higher content of –OH or –COOH on rCF surface in comparison with vCF, where all of these functional groups can be potential sites for forming covalent bonds with the new polymer. Fig. 5 illustrates the surface morphology of fibres recycled through this method. Having a higher IFSS in rCF-based composites is in good agreement with the findings reported by Wada et al. (Wada et al., 2016). Based on their results, pyrolysis of CFRC under N_2 atmosphere and in the presence of superheated steam at 700 °C led to having a rCF composite specimen with IFSS level comparable with that of sized vCF (Wada et al., 2016). The results reported by Ma et al. (Ma et al., 2020) corroborated the role of steam in lowering the weight loss of CF. It was reported that the presence of 10% oxygen and air in the pyrolysis step at 700 °C can lead to around 26% and 37% weight loss of fibres, respectively. This is while in the presence of N_2 and steam, the weight losses were reported to be only about 0.22, and 0.19%. These observations highlight the importance of having a fine control on the influential parameters such as temperature, duration, and reaction atmosphere during the recycling process (Ma et al., 2020).

Microwave-assisted pyrolysis: This is a comparatively new procedure that has been used as a heat source in the pyrolysis process of composite materials (Jiang et al., 2015; Lester et al., 2004). Indeed, it has already been used to recover glass fibres from the composite parts used in turbine blades (Åkesson et al., 2012). Microwave-assisted pyrolysis is considered as an efficient, fast and eco-friendly approach compared with traditional recycling pyrolysis method (Deng et al., 2019; Donaj et al., 2010). In a study conducted by Jiang et al. (Jiang et al., 2015), the pyrolysis process was carried out under nitrogen flow in a microwave furnace at 400, 500, and 600 °C for 30 min and the performance of rCF in developing new composite products was investigated. The obtained results demonstrated that even at the highest pyrolysis temperature, not only a complete cleaning of CF surface was not achieved, but it led to some damages to fibres leaving cavities and defects on their surface. rCF and vCF at the ratios of 0–30 wt% were used to fabricate polypropylene (PP) and nylon composites via extrusion. The results revealed that their mechanical performance depended largely on the type of the matrix, fibre surface morphology and the interfacial interactions between fibres and polymers. For instance, the reinforcing rCF in PP outperformed vCF, while an opposite trend was observed in nylon-based composites. This was due to different interactions between the fibres and matrices and outweighing dominant factors such as surface roughness in rCF (Jiang et al., 2015). Because vCF had a negligible chemical and physical interactions with the non-polar PP matrix, the rough surface of rCF was the dominant factor in increasing the physical interactions with the matrix leading to higher tensile strength and Young's modulus. It is highly recommended to investigate the impact of microwave-assisted pyrolysis method on surface chemistry and elemental composition of fibres as there is gap on this in literature. This will provide a clearer picture on the applicability of the recovered fibres in developing new composite products and structures.

2.2.2. Fluidised bed

Fluidised bed thermal processing is a technique based on which the polymeric matrix can be separated from the embedded fibres using a stream of hot air in the presence of silica sand (Yip et al., 2002; Turner et al., 2011). There are few recent research studies on this recycling process and understating the role of contributing factors in determining the characteristics of recovered fibres. For instance, thorough investigations have been carried out at the University of Nottingham on different aspects of using this method for recycling carbon fibres (Yip et al., 2002). The composite scraps should be shredded into the size range of 6–20 mm before entering into the silica bed. The size of the silica particles is around 850 μm and the process is carried out at 450–550 °C depending on the polymer type using a flow of hot air (Pickering, 2006; F. Meng et al., 2017). Under this condition, the

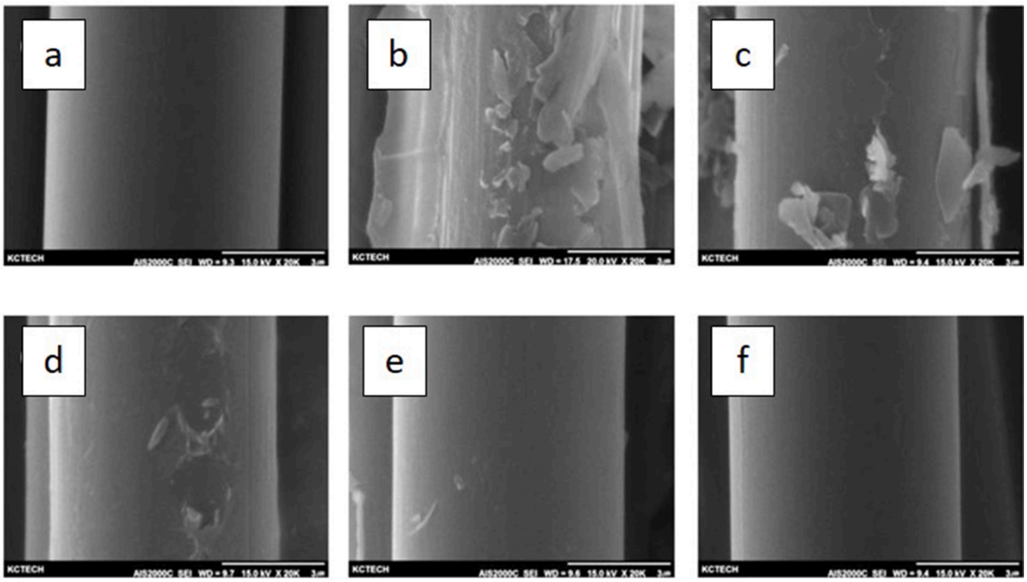

Fig. 5. SEM images of desized vCF, b) CF after steam pyrolysis at 550 °C; rCF after steam/air process treated for c) 30 min, d) 45 min, e) 60 min, and f) 75 min (Reprinted with permission from ref (Kim et al., 2017), Copyright 2017 Elsevier).

polymeric matrix is vaporised, releasing clean fluffy fibres of different lengths and orientations (Yip et al., 2002). The fibres are then transferred to a secondary chamber via the air flow leaving behind other fillers and additives to undergo an oxidation process and get the remaining polymer deposits fully oxidised (Meng et al., 2018; F. Meng et al., 2017). In terms of mechanical properties, the recycled fibres may experience up to 20% reduction in their tensile strength. However, this thermal process has shown limited influence on other aspects of fibres such as stiffness and surface oxygen content. It has been reported that through this recycling process some of the surface hydroxyl groups on CFs can be transformed to carbonyl ($C = O$) and carboxylic groups (–COOH) due to the applied heat. However, the interfacial shear strength with epoxy resin did not change (Jiang et al., 2008). The fluidised bed process has also been utilised to recover glass fibres from the composite wastes; however, the obtained fibres showed a severe depletion in their tensile strength, where up to 50% reduction was observed (Pickering et al., 2000).

The recovered fibres can be used in developing composites using compounding and injection moulding methods in the presence of suitable coupling agents such as maleic anhydride grafted polymers (Wong et al., 2012). Despite all the advantages, the method has mainly been used in the lab scale experiments (Piñero-Hernanz et al., 2008). Shortened recovered fibres length range and low recovery yield of polymers after recycling process are some drawbacks of this method (Marsh,

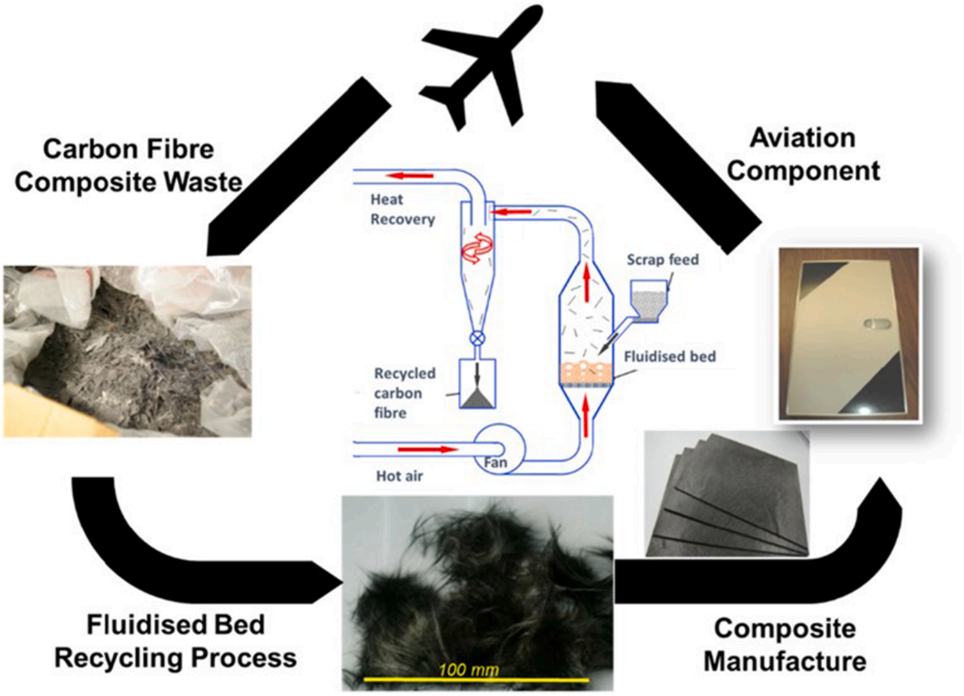

Fig. 6. The schematic illustration of a fluidised bed thermal process and the closed loop of using rCF in aviation industry (Reprinted with permission from ref (Meng et al., 2020), Copyright 2020 Elsevier).

2009). Fig. 6 shows a schematic representation of this recycling process and the closed loop of using rCF in aircraft interior applications.

2.3. Chemical recycling of CF

2.3.1. Supercritical fluids (solvolysis)

The chemical recycling of carbon fibre via solvolysis, involves breaking down the polymeric matrix into soluble lower molecular weight products using different types of reactive solvents (Oliveux et al., 2015). The final products will be fibres, inorganic fillers and dissolved depolymerised resins and monomers (Morin et al., 2012; Oliveux et al., 2015). To this end, near- and supercritical solvents such as water, alcohols, ammonia, organic solvents at different temperatures and pressures have been used where the resin removal efficiency of between 95 and 100% has been reported (Morin et al., 2012). The supercritical fluids have some features such as liquid-like density, gas-like viscosity, dissolving ability, and high diffusivity which all can be helpful in recycling the CFRC (Xu et al., 2013; Okajima and Sako, 2017).

Some of the main advantages of this method are i) obtaining clean fibres without significant deterioration of mechanical properties, ii) maintaining the length of fibres fed to the reactors, and iii) rapid and selective depolymerisation (Goto, 2009). However, this recycling process is expensive since it requires specific types of reactors and facilities capable of operating at high temperatures and pressures, and also in the presence of corrosive media (Oliveux et al., 2015). As a result, the overall industrial readiness of solvolysis has been evaluated to be less than that of thermal and mechanical recycling methods (Khalil, 2018). Among different types of fluids, the efficiency of water and alcohols have widely been studied. Water has mainly been utilised as the fluid in this recycling method due to its nontoxicity, ease of handling and recyclability (Kim et al., 2019). In a supercritical condition, the dielectric constant of water molecules reduces and therefore water acts as a non-polar solvent which can dissolve the organic compounds (Kim et al., 2019). The use of alcohols can be easier because of their lower critical point conditions (200–300 °C and 2.0–6.0 MPa) compared to water (374 °C and 22.1 MPa); however, their environmental effects should be considered (Jiang et al., 2009). The addition of some acidic or alkaline compounds as well as introducing oxygen into the systems have also been effective in accelerating and improving the efficiency of resin removal. Overall, the removal of resins through solvolysis encompasses several steps including i) diffusion of solvent to the fibres surface, ii) reaction of fluid with the fibres surface, and iii) dissolution of matrix into fluid, and iv) mass transfer by convection in the fluid (R. Piñero-Hernanz et al., 2008).

There are several studies on recycling CFRC using solvolysis (Kim et al., 2019; Oliveux et al., 2017; Henry et al., 2016). Ponero-Hernanz et al. (Piñero-Hernanz et al., 2008) studied using near- and supercritical water over a temperature range of 523–673 K (250–400 °C) and pressures of 4–28 MPa in the presence of hydrogen peroxide (H_2O_2). Potassium hydroxide (KOH) was also added as a catalyst to the system and the effects of temperature, solvent concentration, catalyst concentration and exposure time were investigated (Piñero-Hernanz et al., 2008). Based on their results, temperature and treatment time were the most influential parameters in resin removal, while the H_2O_2 concentration negatively affected the overall yield (Piñero-Hernanz et al., 2008). This was due to the low stability of H_2O_2 in the mentioned critical conditions. The presence of KOH significantly improved the resin removal yield with efficiencies of 79.3 wt% and 95.4 wt% in the absence and presence of the catalyst, respectively. In terms of mechanical characteristics, the tensile strength of fibres was reduced by 2–10% compared with vCF (Piñero-Hernanz et al., 2008). Similarly, the synergetic impact of catalysts such as sulphuric acid (Yuyan et al., 2009), KOH/phenol (Liu et al., 2012) and oxygen (Bai et al., 2010) on improving the recycling efficiency of water based systems was investigated. All these additives improved the overall yield, and it was demonstrated that adjusting the content of catalysts into the system was crucial to maximising efficiency. In addition, selecting the appropriate solvent and catalysts type over different temperature ranges was effective in improving resin removal. For instance, Liu et al. (Liu et al., 2012) showed that through using phenol and KOH in recycling solution, the recovery efficiency of fibres reached 95.2 wt% and 100% within 30 min treatment at 315 and 325 °C, respectively. The mechanical characteristics and surface chemistry of recovered fibres were comparable with those of vCF (Liu et al., 2012). The research conducted by Henry et al. (Henry et al., 2016) revealed that the water/alcohol mixture (50/50 vol. %) can lead to a higher resin removal efficiency than pure water. The effects of some parameters such as temperature, duration and solvent were investigated. It was observed that water/ethanol resulted in a cleaner rCF surface which was because of the faster decomposition rate in comparison with pure water. This reduced the chance of forming pyrolysed carbon residues on fibres and lowering the contact time between the resin and carbon fibres which caused forming less residues on the fibres. It was suggested that the shortening of treatment duration can lead to a cleaner fibre surface. Kim et al. (Kim et al., 2019) reported 99.5% resin removal efficiency using supercritical water without any catalysts after optimisation. The recycled fibres were reused and mixed with cyclic butylene terephthalate (CBT) to produce a thermoplastic composite using compression moulding. It was reported that for complete removal of resin, 120 min of treatment with supercritical water was required. Okajima and Sako (Okajima and Sako, 2017) reported that the recycling process using high-pressure superheated steam at 400 °C, 20 MPa, for 45 min, reduced the content of functional groups of –COOH, $-C = O$ and C–OH on rCF compared with vCF. These findings are in contrast with the findings of Bai et al. (Bai et al., 2010) results which demonstrated significantly higher O/C ratio on rCF surface compared with vCF.

In addition to water and mixtures of water, different types of organic solvents such as alcohols have also been used due to their less extreme supercritical conditions and their higher solubility of polar and high-weight molecules (Yan et al., 2016). For instance, methanol, ethanol, 1-propanol, n-propanol and acetone were used in the presence of different types of catalysts (KOH, NaOH and CsOH) over 200–450 °C (Jiang et al., 2009; Piñero-Hernanz et al., 2008). Some influential parameters such as the impact of flow rate and the presence of catalysts on resin removal efficiency were investigated by Ponero-Hernanz et al. (Piñero-Hernanz et al., 2008). It was realised that the resin removal efficiency was altered by changing the temperature, pressure and solvation capability of fluids. For instance, at 300 °C and in lower pressures, acetone and ethanol showed 25.4 wt.% (at ~8 MPa) and 8.7 wt.% (at ~4.5 MPa) efficiencies, but these increased to 78.7 and 78.8 wt% respectively, when the pressure was doubled and the temperature increased to 450 °C. Furthermore, the catalyst improved the efficiency of resin removal from around 10 wt.% to 86 wt.% after adding KOH using 1-propanol at 300 °C during a 15 min treatment. The mechanical agitation of samples was avoided because of its possible damage to the mechanical characteristics of fibres. Therefore, to preserve the mechanical properties of the fibres, a semi-flow system of fluids with a controlled flow rate was recommended instead of mechanical agitation (Piñero-Hernanz et al., 2008). Characterising the rCF recovered through supercritical *n*-propanol in a continuous flow system revealed that the mechanical properties of fibres remained unchanged (Jiang et al., 2009). However, the concentration of surface oxygen of fibres reduced significantly causing some negative impacts on interfacial shear strength with a new matrix (Jiang et al., 2009). These results may vary based on selecting different experimental conditions or solvents. Table 1 lists the summary of some of the research studies published in this arena.

2.3.2. Low temperature chemical recycling methods

Using supercritical fluids requires specific facilities to control temperature and pressure, making it an expensive and energy-intensive process. Therefore, some CFRC recycling research studies have focused on developing easier experimental conditions for chemical degradation

Table 1
Some publications on reclaiming CF from CFRC using solvolysis method.

Authors	Solvent type	Catalysts	Experimental condition	Efficiency (%)	Mechanical Characteristics	Ref
Ponero-Hernanz et al	Water	H_2O_2 and KOH	$T = 523$ to 673 K, $P = 4$- 27 MPa	79.3% (SCW), 95.3% (With catalyst)	Tensile strength: 2–10% reduction Tensile modulus: N/A	(R. Piñero-Hernanz et al., 2008)
Kim et al	Water	None	$T = 405 \pm 2$ °C $P = 280 \pm 10$ bar $t = 10$, 30, 60, and 120 min	90% (after 10 min), 99.5% after (120 min)	Tensile strength: 18–36% reduction Modulus elasticity: 7.2–20.2%	(Kim et al., 2019)
Jiang et al	n-propanol	None	$T = 310$ C $P = 5.2$ MPa $t = 20$ min Flow rate: 2 ml min^{-1}	No resin removal percentage was reported	No significant changes in tensile strength and modulus were reported Lower surface oxygen Lower IFSS	(Jiang et al., 2009)
Piñero-Hernanz et al	Methanol Ethanol 1-propanol Acetone	alkali catalysts (NaOH, KOH and CsOH from 0.016 to 0.50 M)	Batch and semi-continuous systems were used $T = 300$- 450 °C $P = 0.1$ to 30.0 MPa $t = 15.5$ min Flow rates= 0.1- 20 ml min^{-1}	More than 95%	Tensile strength: 1–15% reduction	(R. Piñero-Hernanz et al., 2008)
Henry et al	Water, Water/ethanol 50/50 vol.%	None	$T = 350$, 375, 400 C $P = 25$ MPa $t = 60$ and 120 min	86–100% depending on the experimental conditions	Increasing temperature resulted in a smoother fibre surface.	(Henry et al., 2016)
Liu et al	Water	Phenol/KOH	Phenol/KOH/Water: 10/1/100 $T = 315$ °C, $t = 30$ min $T = 325$ °C, $t = 30$ min	95.2% 100%	Tensile strength: 2.62 GPa (vCF), 2.63 GPa (rCF) Tensile modulus: N/A	(Liu et al., 2012)
Bai et al	Water	Oxygen	$T = 440 \pm 10$ °C $P = 30 \pm 1$ MPa $t = 25$–35 min	94–97%	Tensile strength: 3.11 GPa (vCF), 3.13 GPa (rCF) at fibre recovery efficiency of 96.5%	(Bai et al., 2010)
Yan et al	1-propanol	KOH	$T = 260$–340 °C $t = 30$–180 min	–	Increasing temperature and treatment time reduced the tensile strength of fibres Higher surface oxygen content was reported	(Yan et al., 2016)
Knight et al	Water	KOH (0.05 M)	$T = 410$ °C $P = 28$ MPa $t = 15$, 30, 120 min	95.9% (after 15 min), 98.6 (after 30 min), 99.2% (after 120 min)	Short fibres with smooth surface. No significant change in tensile strength after 120 min treatment	(Knight et al., 2012)
Keith et al	Acetone/water 80/20 vol.%	None	$T = 300$–380 °C $P = 16$–30 MPa $t = 150$ min	up to 95 wt.%	Damages to the woven texture were observed at $T > 340$ °C	(Keith et al., 2019)
Ibarra et al	Water, Benzyl alcohol	None	$T = 400$ °C $t = 60$ min	89.1% with supercritical water 93.7% with supercritical benzyl alcohol	Microspheres resulted from the resin decomposition were formed on fibres. No data on mechanical properties were reported.	(Morales Ibarra et al., 2015)
Okajima et al	Methanol	None	$T = 270$–350 °C $P = 5$, 8, 10 MPa $t = 10$ min-90 min	–	Tensile strength decreased by 9% IFSS decreased by 20% rCFRC was produced which had almost equal interlaminar shear strength almost equal to that of vCFRP	(Okajima et al., 2014)
Yildirir et al	Ethylene glycol (EG), and EG/water	None	For pure EG: $T = 300$–390 °C $P = 4.2$ MPa $t = 0$ and 10 min, Fr EG/water: $T = 380$, 400, and 420 °C $t = 0$ min	Up to 89.7% with pure EG, 97.6% with EG/water (5:1)	Resin removal increased with increasing ethylene glycol/water ratio No decrease in the tensile strength With EG/water, the Young modulus decreased by 30%	(Yildirir et al., 2014)

of CFRC, utilising pre-treatments, acids or other types of solvents along with catalysts (Kim et al., 2020). This group of experiments was mainly based on using oxidising chemicals in combination of heat, catalysts, acids, and swelling agents, amongst others. There are however, concerns on the applicability of each of these methods for industrial scale applications. Pre-treatment is usually used as an approach to induce swelling of the composite structure and facilitate interactions between catalysts or oxidising agents. The catalysts should diffuse into the cross-linked network, breaking down the chemical bonds in cured resins (Liu et al., 2017). Strong acids and alkali media have widely been used for their ability to promote chemical degradation of the network. For instance, Jiang et al. (Jiang et al., 2017) reported that after immersing the CF/epoxy resin composites into a nitric acid/sulphuric acid mixture followed by a treatment with polyethylene glycol (macrogol 400) in the presence of KOH, CF was released (Fig. 7). Two parameters including temperature and treatment time were effective on recycling process of fibres where treating the samples at 160 °C for 200 min led to the resin decomposition rate of 97%. This process introduced polar groups such as carboxyl and carbonyl groups onto the carbon fibre surface but also caused around 4% reduction in the tensile strength compared with vCF (Jiang et al., 2017).

Oxidising chemicals such as H_2O_2 have also been reported effective

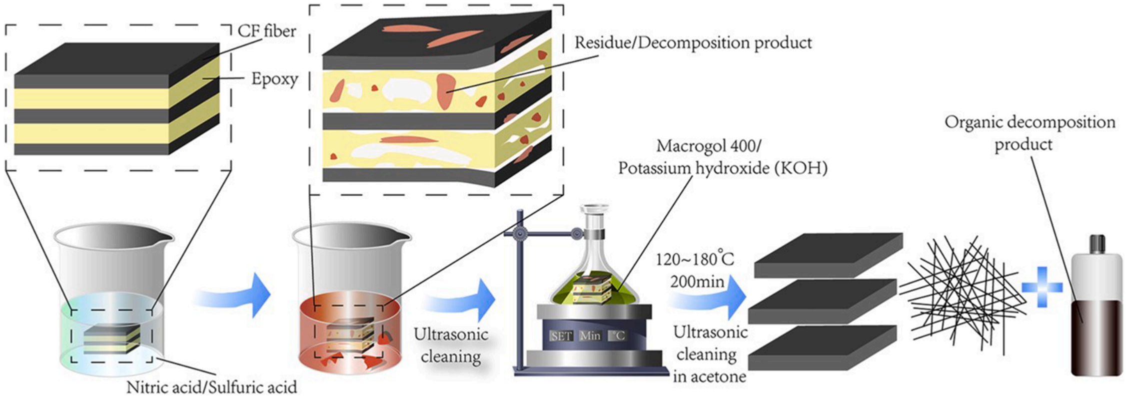

Fig. 7. Recovery of CFs using nitric acid and macrogol 400 (Reprinted with permission from ref (Jiang et al., 2017), Copyright 2017 Elsevier).

in triggering the degradation process of thermoset polymers. H_2O_2 generates hydroxyl radicals which are strong oxidising species. However, H_2O_2 alone seems to have a low efficacy and therefore more is required to improve the resin degradation, such as additional heat or other catalysts (Li et al., 2012). For instance, the CF/epoxy composites were first refluxed with acetic acid at 120 °C and then treated in a H_2O_2 solution containing N,N-dimethylformamide (DMF) (Fig. 8) (Xu et al., 2013; Wang et al., 2015). Pre-treatment process with acetic acid resulted in swelling the composite samples hence a better interaction with the oxidising agent. Next, 0.2 g of pre-treated composites were immersed into the H_2O_2/DMF solution ($<$ 30 mL) in a hermetic autoclave and heated at 80–150 °C for 5–120 min (Xu et al., 2013). Under this condition, H_2O_2 generated powerful oxidising species like hydroxyl radicals (OH) which oxidised organic compounds. The decomposed organic products were then dissolved in DMF and separated. It was realised that optimising time, temperature and volume ratio of DMF/H_2O_2 were critical to control the overall decomposition rate of the resin. An efficiency of 99.1% was obtained at temperatures above 90 °C, 30 min treatment and V_{DMF}/V_{H2O2} 1:2 (Xu et al., 2013). When treated at 100 °C, rCF retained 98% of its tensile strength, while increasing the temperature to above 120 °C, lowered its tensile strength to below 90% (Xu et al., 2013).

Zabihi et al. (Zabihi et al., 2020) accelerated the impact of H_2O_2/TA (tartaric acid) solvent treatment using microwave irradiation and reported synergistic effects in recovering CF from the waste composite panels (WCP) (Fig. 9). This was carried out in the presence of different ratios of H_2O_2/TA (1 to 3) over 1–3 min microwave irradiation periods. The results showed the matrix decomposition yield of up to 95%, the tensile strength retention of 92% and negligible reduction in modulus (Zabihi et al., 2020). In another study, the potential effect of sonochemical processes on decomposing the CFRC was examined by Das and Varughese (Das and Varughese, 2016). Ultrasonication in liquids resulted in enhancing the reaction rates through creating the cavitation phenomenon, high energy release from the implosion of microbubbles, localised temperature of 5000 K (4727 °C) and pressure of 1000 atm, and generating radical species like $H^\bullet$, $^\bullet OH$, and $HOO^\bullet$ from water molecules (Das and Varughese, 2016). The treatment of CFRC with a solution of dilute nitric acid and hydrogen peroxide under ultrasonication led to the degradation of resin by up to 95%, which was three times higher than that without sonication (Das and Varughese, 2016). The duration of treatment was effective where the resin decomposition was noticeable after at least 4 h, and through increasing the time to 8 h, the decomposition rate increased significantly. Comparable tensile strengths with vCF were reported for recovered fibres through this method (Das and Varughese, 2016). It is however recommended to shorten the ultrasonication period and explore new solvents and treatments for higher efficacy of this recycling technique.

Solvents and catalysts are effective in determining the swelling rate of resin and its removal efficiency. Acetic acid generates far higher swelling rate in composites compared with alcohols where the swelling ratios for acetic acid and ethanol were ~50 and ~20%, respectively (Fig. 10b) (Wang et al., 2015). Using acetic acid along with $AlCl_3$ as catalyst at 180 °C over 6 h treatment in a Teflon-sealed autoclave led to an effective recovery of CF from cured epoxy composites (Wang et al., 2015). It was reported that in the presence of 15 wt% $AlCl_3$, 100% resin removal efficiency was obtained (Fig. 10c-f). Similarly, the combination of zinc chloride ($ZnCl_2$)/ethanol in a pressure vessel has been reported effective in recycling aerospace CFRP waste (Tg$>$ 200 °C) (Liu et al., 2017). Liu et al. (Liu et al., 2017) examined the efficiency of different catalysts such as KOH, nitric acid, $ZnCl_2$, $AlCl_3$, and $FeCl_2$ and reported that the $ZnCl_2$ was more efficient in breaking down the CFRC. Ethanol was also preferred to water as solvent due to inducing more swelling to the CFRC and enhancing the interactions between catalysts and chemical bonds. Their results showed that the limiting requirements for resin degradation were 5 h treatment at 190 °C in the presence of 20 wt% catalyst (Liu et al., 2017). No information on the impact of this recycling process on mechanical characteristics of rCF was provided.

The discussed chemical treatments have also been used to recover CF from thermoplastic matrices (Tapper et al., 2018). Tapper et al. (Tapper et al., 2019) reported a closed-loop manufacturing and recycling process for thermoplastic CFRC made of chopped CF (3 mm) and PA6 polymer using the chemical method. The alignment of fibres in composite production process was achieved based on using the HiPerDiF method. This technique aligns discontinuous carbon fibres and involves acceleration of the suspension containing CF through a nozzle, aligning fibres transversely on an orientation plate then removing the fluid and drying

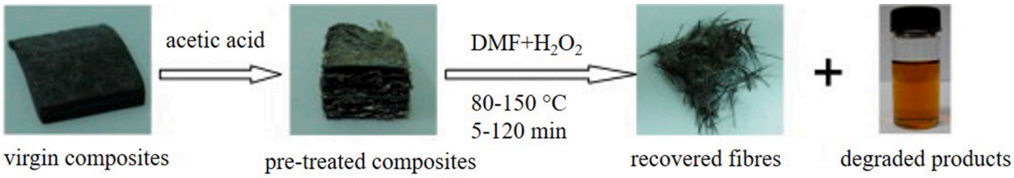

Fig. 8. The chemical decomposition of epoxy resin using acetic acid-H_2O_2-DMF combination (Reprinted with permission from ref (Xu et al., 2013), Copyright 2013 Elsevier).

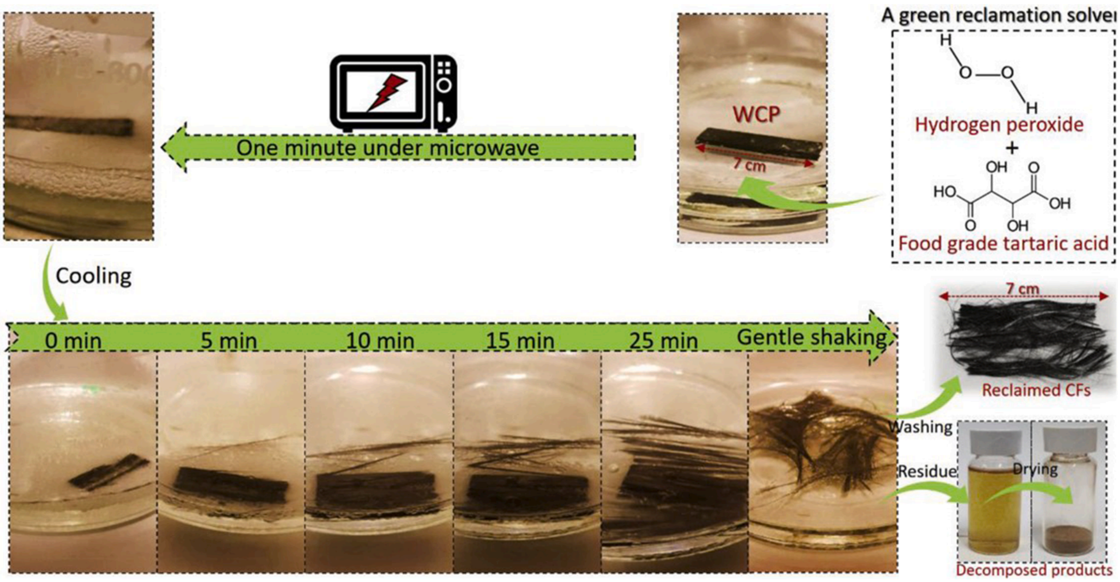

Fig. 9. Microwave assisted recovery of CF from waste composite parts in the presence of H_2O_2/TA as solvent (reprinted with permission from ref (Zabihi et al., 2020), Copyright 2020 Elsevier).

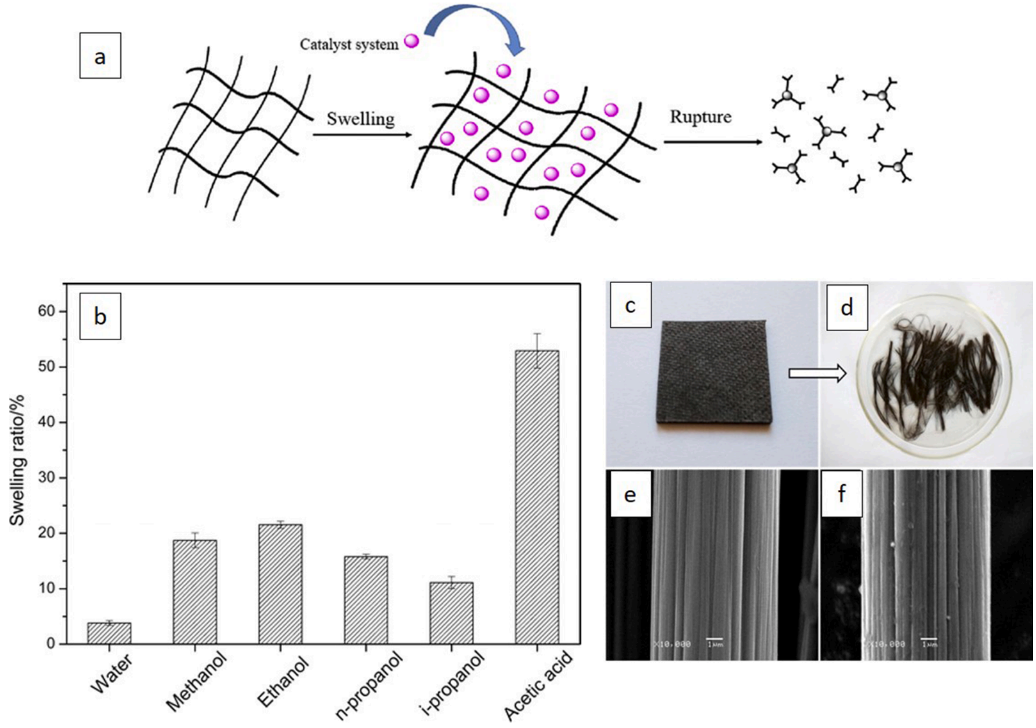

Fig. 10. a) Schematic illustration of the effect of catalyst in chemical degradation of CFRC (Liu et al., 2017) (Reprinted with permission from ref (Liu et al., 2017), Copyright 2017 Elsevier) (b) Swelling effect of different solvents on epoxy composite, c) cured epoxy composite, d) recycled fibres using $AlCl_3$/acetic acid, e) vCF, f) rCF using $AlCl_3$/acetic acid (15 wt.% catalyst, 180 °C, 6 h) (Reprinted with permission from ref (Wang et al., 2015), Copyright 2015 American Chemical Society).

(Yu et al., 2014). Based on the reported work, the CF and PA6 polymer were reclaimed from CF/PA6 composites after treatment with benzyl alcohol at 160 °C for 1 h followed by a filtration. The separated constituents were used once again to manufacture a new CF/PA6 composites and two consecutive recycling loops were carried out. Analysis of the manufactured composites showed a 25 and 39% reduction in tensile stiffness and tensile strength, respectively after first loop of recycling process. The reduction in tensile strength was referred to the fibres agglomeration and misalignment in the manufactured composites. The second time recycling did not affect the tensile strength of composites but reduced the stiffness by further 20% (Tapper et al., 2019).

3. CFRC recycling from energy and environmental benefit perspectives

The main aim of the aforementioned recycling methods is basically increasing the recycling efficiency and maintaining the original characteristics of CFs. Achieving this will augment the applicability of recycled CFs in different new fields and complying with new governmental directives (Witik et al., 2013). Analysing the environmental impact and cost efficiencies is required to obtain a better understanding on the true potential of different recycling methods. In general, it has been demonstrated that the energy consumption of a recovery process of CF from CFRC is much less than that of producing vCF. This is referred as one of the main benefits of recycling CFRC to reclaim the fibres. However, the real environmental impacts of a recycling process can only be assessed by considering all the stages involved (Witik et al., 2013). An environmentally viable recycling method should provide less overall environmental impacts than the combined impacts from the production process of virgin material and other waste management methods (Witik et al., 2013). Therefore, a complete environmental assessment should consider all various factors involved in a recycling chain such as collection of waste, labour cost, governmental legislations, transportation, pre-treatment, recycling, and remanufacturing the recovered materials to provide a more comprehensive picture of the applicability of a recycling process (Witik et al., 2013). LCA and life cycle cost (LCC) analyses are employed as standard methods to assess the environmental and financial benefits of CFRC recycling methods. They are well-established approaches which quantify the environmental and economic aspects of products considering each stage of their manufacturing process. In literature, the environmental benefits of different recycling routes have mainly been compared with landfilling and incineration. The assessments have been carried out on hypothetical scenarios and inventory analyses based on the databases of governments and industries or expert assumptions in different geographical areas. The environmental impacts can generally be assessed based on some metrics such as global warming potential (GWP) and primary energy demand (PED) in LCA models. It is highly likely that the recycling processes result in down-cycling of the recovered materials, which can subsequently affect their potential applications and even type and content of the target virgin material which can be replaced. Therefore, understanding the technical properties of recovered CF from each recycling technique is required to map the future target markets.

Based on a comparative LCA assessment conducted by Dong et al. (Vo Dong et al., 2018), regardless of recovery of energy and materials from the processing of wastes and without imposing any extra cost by governmental regulations, incineration and landfilling have the lowest operating costs compared with other recycling methods. This factor along with the lack of clear market outlook for the recycled materials makes the recycling of CFRC a costly procedure and an unattractive option. Among different methods, the mechanical recycling shows the lowest processing cost. However, this recycling method suffers from some hurdles such as generating low value products and causing severe damages to the fibres, both of which limit the application of the recyclates. Using supercritical water (SCW) or pyrolysis methods for recycling CFRC has shown higher operating costs compared with mechanical recycling (Vo Dong et al., 2018). This is because of their energy intensive nature, the requirement of high investments, and for the case of supercritical fluids the low processing rate. The energy requirement for using microwave pyrolysis was found to be one third of the conventional pyrolysis, and thus it significantly reduced the operating cost (Vo Dong et al., 2018). The supercritical water treatment provided the highest valued fibres; however, the low processing rate was mentioned as a barrier for a broader application of this method (Vo Dong et al., 2018). Fig. 11a compares the efficiency of different recycling pathways based on three indices of operating cost, associated costs charged by governments and industries (UCW), and the value of the final rCF (UCF).

The environmental impacts of each of these recycling technologies were assessed based on their overall GWP, by considering two important factors including GWP of the recycling process (GWPP) and the avoided environmental impact after substituting the target virgin materials (GWPA) with the rCF (Fig. 11b) (Hagnell and Åkermo, 2019). The total environmental impact (GWPTOT) of the studied recycling methods was calculated based on the net difference of GWPP and GWPA. The low

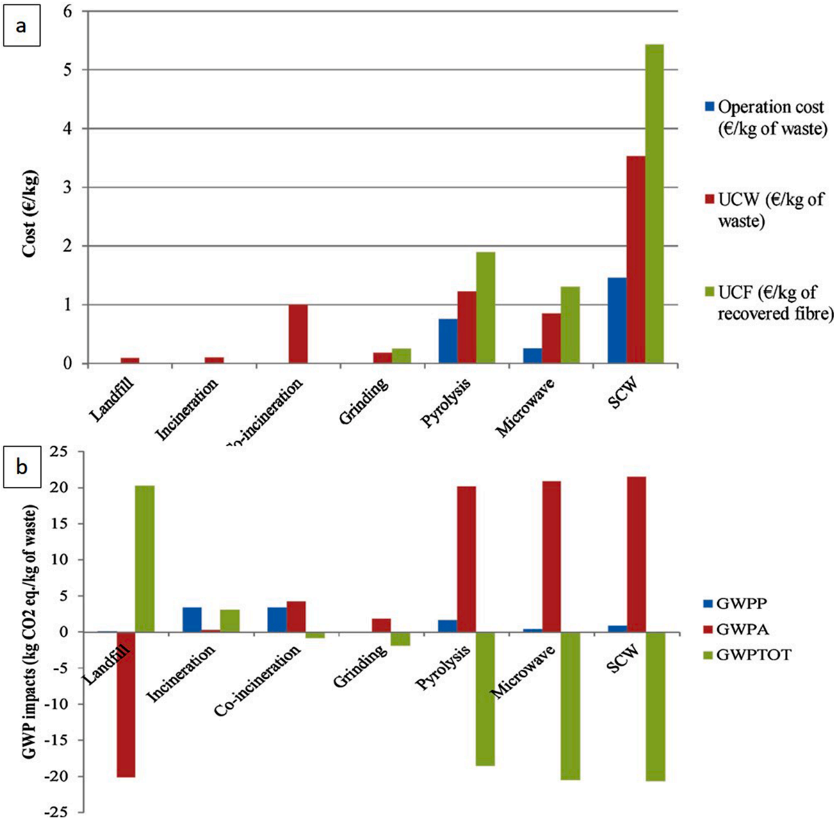

Fig. 11. Comparison of a) economic assessment and b) environmental impact of various waste management and recycling methods (Reprinted with permission from ref (Vo Dong et al., 2018), Copyright 2018 Elsevier).

value of the recycled materials from the mechanical processing led to a relatively low GWPA index, showing the low potential of the recyclates in displacing the virgin materials. In contrast, the pyrolysis, microwave and SCW methods showed higher GWPA values which were due to the recovery of the collected by-products and the applicability of the recovered CF in manufacturing the composite structures. This mitigates the necessity of using vCF and therefore provides a higher environmental impact avoidance after using rCF. Similarly, another analysis conducted by Princaud et al. (Prinçaud et al., 2014) in the context of Europe, corroborated the lower environmental impact of the solvolysis approach using supercritical water in recycling 1 kg of CFRC waste compared with that of landfilling approach.

However, it is worth mentioning that all these findings are subjected to the inventory analysis of models and author assumptions for assessing different impacts of the recycling methods. For instance, in a research conducted by Witik et al. (Witik et al., 2013) based on some assumptions in the Western Europe, the environmental impact of the pyrolysis method was pretty much affected by the final application of rCF. The results showed that the environmental impact of the pyrolysis process compared with incineration and landfilling was influenced by the type of the virgin material, i.e., vCF or vGF, which was supposed to be displaced by rCF. Considering the replacement of vGF with rCF in the manufacturing process of composite parts for automotive industry, it was realised that the pyrolysis recycling method provided greater deleterious environmental impact than landfilling and incineration. However, an opposite trend was observed in case of replacing vCF, which was due to the energy intensive production process of vCF. Therefore, using rCF can significantly lower the potential emission of GHG, resulting in a lower environmental impact.

The viability of mechanical recycling method from economic and environmental point of views has also specifically been examined. However, it is worth mentioning that there are limited studies with up-to-date data on analysing the true potential of the mechanical recycling method. The low performance of mechanical processing in producing high value products and its low energy efficiency can be compensated for by increasing the production rate and capacity. Howarth et al. (Howarth et al., 2014) developed a bottom-up modelling approach to evaluate the energy efficiency of recycling CFRC based on mechanical milling in an industrial scale. Their study demonstrated that the mechanical processing of wastes could be more energy-efficient at higher recycling rates and throughputs, where the processing energy of 2.03 and 0.27 MJ/kg were calculated for 10 and 150 kg/h waste processing rates, respectively. The calculated processing energy was much lower than the embodied energy of vCF (183–286 MJ/kg) showing the economic and environmental benefits of this recycling method. The limited potential applications of rCF recovered using mechanical approach should also be highlighted, as a factor which can significantly affect the justification of this recycling method.

The environmental benefits and financial performance of mechanical recycling of CFRP obtained from end-of-life vehicle components were assessed by Li et al. (Li et al., 2016) based on LCA and LCC models and the outcomes were compared with landfilling and incineration methods. The assessment was conducted based on the waste management regulations of UK and EU. Landfilling had the least processing cost amongst the examined methods, but it had zero contribution to energy recovery from CFRC waste. Incineration showed the least energy demand for processing the CFRC waste because of its high contribution to returning energy in the form of heat and electricity; however, it had the highest GHG emission compared to other waste treatment methods. In respect to mechanical recycling, it was environmentally beneficial only at certain conditions by lowering the GHG emission. Its environmental benefits were justifiable if rCF could displace vGF in composites manufacturing process, provided that the remaining course recyclates be landfilled instead of incinerated. However, the high processing cost, resulting mainly from dismantling and recycling processes, low value of rCF, and low revenue of target markets, was highlighted as a barrier to the financial viability of this waste treatment method (Li et al., 2016). The LCA and LCC analyses are therefore subjected to selected parameters and costs and as a result, the reported findings vary depending upon assessment boundaries and location (Li et al., 2016).

The environmental benefits of fluidised bed recycling method has also been investigated in some studies (Meng et al., 2017; Meng et al., 2017). The feed rate of CFRC per unit area of fluidised bed is an effective factor in determining the energy efficiency of the recycling process and also the final length of fibres (Meng et al., 2017). It has been reported that the energy efficiency of recycling CF through this method can be 3–10% of the energy required for production of vCF (Meng et al., 2017). In an investigation conducted by Meng et al. (Meng et al., 2017), the fibres reclaimed using fluidised bed method were used to manufacture rCFRC for the automotive industry through compression moudling, injection moudling, and autoclave moudling. Two environmental metrics including PED and GWP were evaluated in comparison with other conventional materials such as steel and aluminium. Their findings demonstrated a major reduction in the GWP and PED after using rCFRC in automotive components which was mainly due to the reduction in weight-induced fuel consumption, lower embedded energy of materials, and the energy efficiency of the employed recycling method. Analysis showed that the manufacturing method of rCFRC components also affected the overall environmental impact of this recycling method. For instance, rCFRC manufactured through compression moudling and autoclave moudling showed much less contribution to the GWP than injection moudling. This was due to the flexibility of these methods in handling high contents of rCF, and therefore producing lighter-weight products. The final application of the rCFRC was also critical as it impacted the overall weight reduction in a vehicle. For instance, the bonnet made of rCFRC had more effect on weight saving than a window frame. Therefore, in this case, the final application of recycled materials along with the manufacturing method were effective in determining the final GWP and PED and understanding the true potential of rCF in replacing conventional lightweight metals such as aluminium.

4. Recycling methods for dry carbon fibres waste

In addition to CFRC scrap, another common type of waste is the dry fibre produced during the manufacturing stages. These waste fibres are from offcuts, bobbin ends and sometimes selvedge. It is estimated that dry fibres account for around 40% of the total CF waste, and because they have not been embedded in any polymeric matrix yet, they possess the identical characteristics of vCF (Badrul Hasan et al., 2019). The production of this amount of valuable waste has encouraged researchers to develop new recycling strategies such as spinning CF into yarns, fabricating nonwovens, and non-crimp prepreg fabrics using combined recycled and virgin CF.

4.1. Spinning rCF hybrid yarns

The methodologies for spinning CF yarns have been developed based on traditional textile spinning methods. The process of making yarns starts with carding process, which is necessary for aligning fibres, eliminating entanglements, and removing some short fibres (Khurshid et al., 2019). The generated card webs are then transformed into slivers and then passed through the drafting, drawing, and spinning stages to achieve spun rCF yarns. However, the waste dry CFs are mainly in the form of staple fibres and due to their intrinsic brittleness, low elongation, low shear resistance and lack of natural crimp, new arrangements should be applied for their processing (Hasan et al., 2018; Hengstermann et al., 2016). These properties make it very difficult to spin the pure carbon fibres into yarns; so blending with thermoplastic fibres such as PA6, PP and polyester has been utilised to obtain hybrid yarns. The added fibres provide support for the staple CFs and enhance the fibre-to-fibre cohesion to provide better CF alignment during the carding process. Moreover, it is required to optimise the parameters of the carding machine such as the clearances between different rollers and also doffer wire-clothing based on the type and length of fibres (Khurshid et al., 2019).

A comprehensive research on spinnability of the CF hybrid yarns has been carried out in the Institute of Textile Machinery and High Performance Material Technology (ITM) of TU Dresden University in Germany. The research mainly focused on fabricating hybrid yarns of CF/PA6 and understanding the impact of influential parameters at different stages of manufacturing process (Badrul Hasan et al., 2019; Hengstermann et al., 2016). The roles of three main parameters including the mixing step prior to carding, CF fibres length and volume, and sizing effects on processability, visual and mechanical characteristics of CF/PA6 fibres were investigated (Hengstermann et al., 2016; Hengstermann et al., 2016; Hengstermann et al., 2019). The results demonstrated that the physical appearance and mechanical properties of the developed yarns are greatly influenced by the length and final content of carbon fibres in the yarn.

The first step in preparing the hybrid yarns is to combine the CF waste with carrier PA6 fibres. In this step, two parameters of i) the initial length of CFs and also the ii) mixing method, determined the final shortening of CFs in the yarns (Hengstermann et al., 2016). The initial length of fibres was selected as 40, 60, 80, and 100 mm and their mixing with PA6 60 mm fibres (50% volume) with and without compressed air mixing was investigated. The fibre bundles were fed into the carding machine to achieve the carding web followed by drafting process using the draw frame to produce hybrid slivers (3.5 ktex). The CF/PA6 yarns (800 tex) were spun using the flyer spinning method. In addition, the prepared yarns were turned into composites after wrapping around a frame and heating at 280 °C for 30 min. The results indicated that the air mixing process led to more homogeneous slivers and yarns due to the intermingling process. However, this mixing caused more damage to the CFs, leading to the overall shortening of their length in the yarns. Analysing the mechanical properties of yarns showed that yarns prepared without air mixing process had a higher tenacity which was attributed to the effect of preparation step in breaking the fibres and shortening their overall length (Hengstermann et al., 2016). The initial length of fibres was found to be detrimental to properties of yarns since it influenced on the final length of the processed fibres in the yarns structure. For example, the longest mean fibre length was observed in yarns prepared with 80 mm CF, which justifies the observed higher tenacity (~23 cN/tex) for the yarns (Hengstermann et al., 2016). Similarly, the unidirectional (UD) composites from yarns made of 80 mm CF and PA6 showed a tensile strength of 800 MPa which had the highest value among other prepared yarns (Hengstermann et al., 2016).

The impact of initial fibre length and mixing ratios on the carding processing and characteristics of developed yarns was further investigated by Hengstermann et al. (Hengstermann et al., 2016). Carding of fibres was carried out after modifying the carding machine parameters including the distances between different carding rollers, and the card wire clothing specifications (Hengstermann et al., 2016). In particular, the distances between Worker-Cylinder, Stripper-Cylinder, Take in-Cylinder and Cylinder-Doffer in the carding machine were adjusted (Fig. 12). The CFs and PA6 fibres in two lengths of 40 and 60 mm were blended manually based on 30, 50, and 70% volume ratios. The carding produced CF/PA6 webs which were then sent to the drawing step to produce slivers (Fig. 13), followed by the flyer machine spinning to make the hybrid yarns. In processing the prepared webs, it was also required to modify the drafting and spinning parameters such as feeding rate, drafting ratio, rollers materials, and twisting numbers to minimise the possible damage to CFs in the slivers (Hengstermann et al., 2016). The results demonstrated that the initial length of fibres and the content of CF both significantly affected the final characteristics of carding webs, slivers and yarns. For instance, the yarns generated from 60 mm CFs showed higher orientation, less hairiness, higher tenacity and less elongation compared with the yarns generated from CF 40 mm in length. This was due to the better carding alignment of longer fibres, causing less damage and less loss of fibres. This in turn increased the overall content of CFs in the slivers and then yarns, which directly contributed to the mechanical characteristics of the yarn. Also, the presence of longer CF fibres, as well as higher volume of PA6 in the slivers resulted in a higher fibre-to-fibre cohesion force and better spinning of slivers (Hengstermann et al., 2016). The orientation of CFs in yarns, the length of CFs and also the twist level of the yarns all were found effective in the final tensile strength of UD composites (Hengstermann et al., 2017). In general, an inverse relation was established between the fibre length and yarn twist with the overall strength of the developed composites. This was due to their impact on the final content and length of CFs in the produced composites as well as the polymer infiltration during the hot pressing process (Hengstermann et al., 2017).

In another work, the impact of CF sizing on the spinning process of yarns was investigated by Hengstermann et al. (Hengstermann et al., 2019). The sizing of CF affected the carding process of fibres through three mechanisms: i) preventing the fibres bundle diameter reduction during carding, ii) protecting the bundles against the applied bending

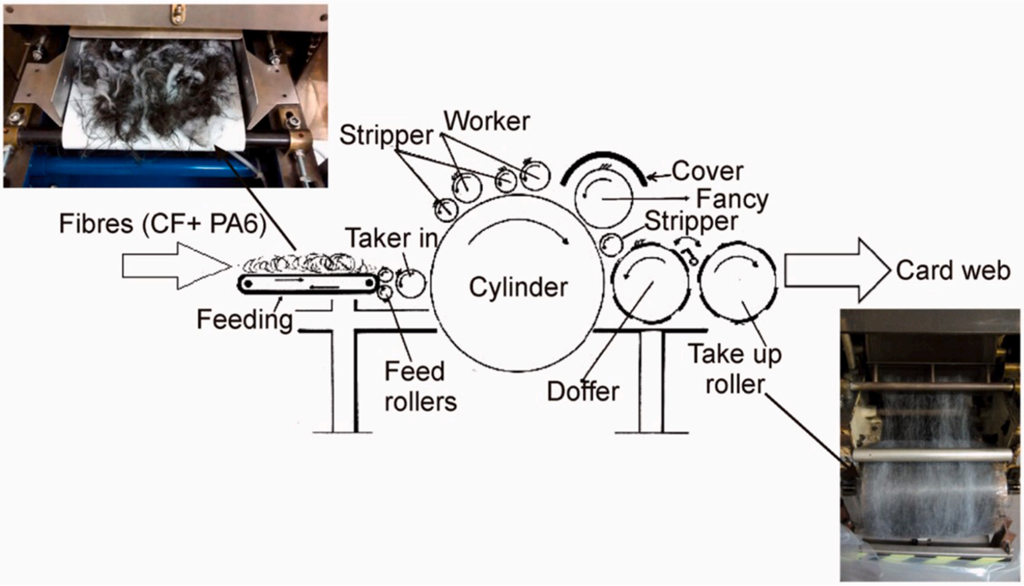

Fig. 12. The carding process for preparing rCF/PA 6 blends (Reprinted with permission from ref (Hengstermann et al., 2016), Copyright 2015 SAGE).

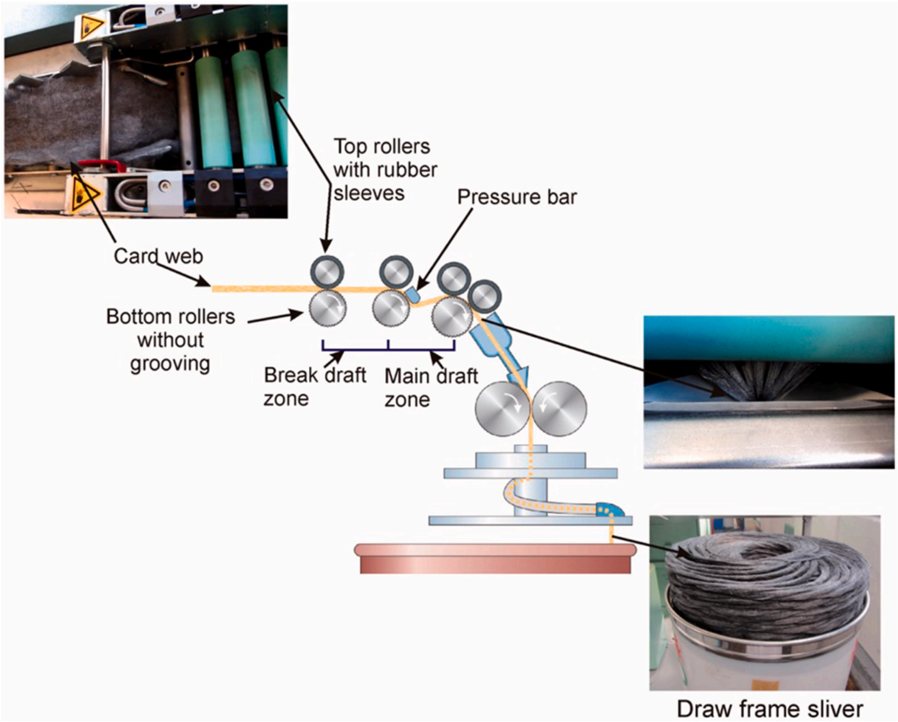

Fig. 13. Drawing process for making slivers from rCF/PA6 webs (Reprinted with permission from ref (Hengstermann et al., 2016), Copyright 2015 SAGE).

forces, and iii) decreasing the friction forces between fibres through smoothening their surface (Hengstermann et al., 2019). The type of sizing also affected the length of CFs after the carding process where the CFs with thermoset-epoxy (CF-E)-compatible sizing had a higher average mean length than the thermoplastic one (CF-T). In addition, the type of sizing was an important factor in determining the mechanical characteristics of unidirectional (UD) composites. For instance, it was observed that the UD composite strips made of CF-E/PA6 and CF-T/PA6 yarns had tensile strengths of 1110 MPa and 990 MPa, respectively, while this value for the samples made of fibres without sizing was 843 MPa (Hengstermann et al., 2019).

Akonda et al. (Akonda et al., 2012) used thermoplastic PP fibres in producing CF hybrid yarns. They produced hybrid rCF/PP yarns using modified carding and wrap spinning processes. To this end, 50–55 mm rCFs recovered from a thermal recycling process of CFRC were mixed with 60 mm PP fibres based on the weight ratios of rCF:PP 30:70 and 50:50. The slivers obtained through carding and then drawing, were wrap-spun into hybrid yarns (Fig. 14a-c) (Akonda et al., 2012). The drawing step was crucial to align the rCF and improve the mechanical properties of the yarn (Miyake and Imaeda, 2016). Similar to the CF/PA6 yarns, the rCF/PP yarns were wrapped around a frame and then heated at 220 °C, under 20 bar of pressure for 15 min to fabricate the UD composite. Analysing the mechanical features of fabricated yarns indicated that the ratio between rCF and the matrix fibre was an influential factor since including a higher initial content of rCF led to more fibre loss during the carding process (Akonda et al., 2012). This was mostly due to the effect of processing on breakage of fibres and low fibre-to-fibre cohesion forces. In addition, after the carding process, the mean length of rCF reduced from 55 mm to 23 ± 3 mm and 17 ± 3 mm in 30:70 and 50:50 blended yarn samples, respectively (Akonda et al., 2012). The presence of more and longer fibres in the yarn structure enhanced mechanical properties and strengthened the developed yarns. Therefore, the composites made of 50:50 slivers showed higher mechanical properties compared with 30:70 samples (Fig. 14d-g) (Akonda et al., 2012). The non-polarity of PP surface also reduced the overall interfacial strength of the composites where rCFs were pulled out during the testing (Fig. 14h and i). Hasan et al. (Hasan et al., 2018) also produced core-sheath CF/PA6 yarns using friction spinning where PA6 fibres were introduced as sheath to CF/PA 6 (62.5% carbon fibre) drafted slivers in the spinning process.

In order to lower any further damages to CF during the spinning process, Xiao et al. (Xiao et al., 2019) reported developing card web carbon fibre reinforced thermoplastic (CWTs) sheets which can then directly be used in moudling process (Fig. 15). In this strategy, CF waste with the length of 60 mm was blended with PA fibres with core-shell structure where the shell was made of polyamide 6 (PA6)–polyethylene (melting point 136 °C) copolymer and the core was polyamide 66 (PA66). The blends of CF and PA fibres were first mixed with the ratios of 20%, 30%, and 40% CF volume fraction and then carded to develop carding webs. The developed webs were then stabilised through stretching and calendar pressing at 110 °C where the outer layer of matrix PA fibres were melted and generated some bonding structures throughout the web, producing CWT sheet. It was realised that the stretching process (30- 60%) contributed significantly to the better alignment of staple CF in the structure of CWT sheets. The developed sheets were used in panel moudling using compression moudling method at 280 °C under pressures of 5–9 MPa. The tensile modulus up to 45.6 GPa was recorded for the compressed sheets. Increasing the content of CF in the CWTs increased the tensile strength and modulus levels. In addition, increasing the stretching ratio contributed to improving the tensile strength mainly in the longitudinal direction.

4.2. rCF-based nonwovens and prepregs

Another route for recycling waste carbon fibres is to use them in the production of nonwoven webs. This method provides great potential for recycling and reusing waste carbon fibres in high value added products (Ghossein et al., 2018). The EGL Carbon Fibre company in the UK has industrialised and optimised the production of rCF nonwoven mats with the capacity of 250 t/year for potential use in the automotive industry (Fig. 16).

Based on the fibres initial lengths, different methodologies such as wet laying and dry laying can be employed. However, very few research studies have focused on developing nonwovens made of rCF (Wölling

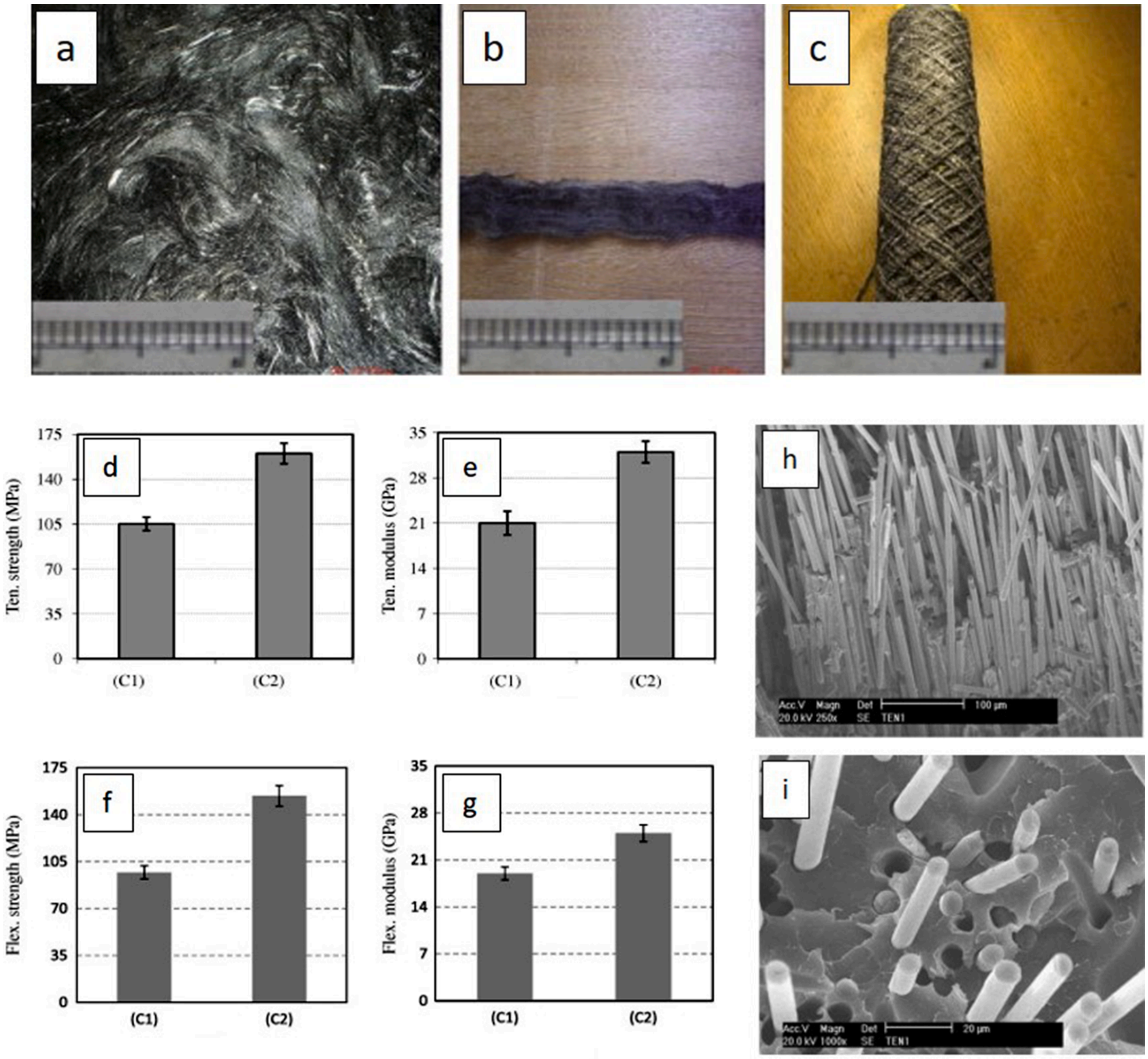

Fig. 14. a) rCF/PP blends 50:50, b) the rCF/PP sliver, c) rCF/PP yarn, D-g) mechanical characteristics of hot-pressed rCF/PP composites (C1: 30:70, C2: 50:50 blends), SEM images of h) aligned rCF in the hot-pressed rCF/PP composites, i) rCF pulled out from the PP matrix (Reprinted with permission from ref (Akonda et al., 2012), Copyright 2011 Elsevier).

et al., 2017). The production of hybrid nonwovens containing rCF and other types of fibre such as PP and flax has been reported (Wölling et al., 2017; Tse et al., 2018). In one study by Wolling et al. (Wölling et al., 2017), the dry-laid and wet-laid nonwovens were prepared and their performance in manufacturing thermoset and thermoplastic composites were compared. The dry-laid samples were produced through carding 100% pure rCF (5–100 mm in length) or its blend with thermoplastic PP fibres, followed by cross-lapping the webs and then needle punching. Two grammages of nonwovens including 250 and 150 g/m^2 were produced through controlling the number of cross-lapped layers (Wölling et al., 2017). In the wet-laid processing, the pure rCF fibres (10–30 mm in length-without sizing) or rCF/PP fibres were dispersed in water and then formed as a sheet on a continuous sieve after eliminating the water. To improve the dispersibility of fibres in solution and handling of nonwovens, carboxymethyl cellulose (CMC) was added to the mixture. In the next stage, the nonwovens were used to manufacture the thermoplastic and thermoset composites via two methods of compression and transfer moulding (RTM) process with epoxy (Wölling et al., 2017). Two interesting findings were reported in this research. First, it was found that the thermoplastic composites made of rCF/PP nonwovens had lower overall mechanical properties than the thermoset composites. This was due to having a sizing layer on fibres which was in nature more compatible with thermoset epoxy resins rather than the thermoplastic PP matrix (Wölling et al., 2017). Second, it was concluded that the composites made of dry-laid nonwovens had higher mechanical properties comparted with those prepared from wet-laid samples. This was due to the role of the carding process in aligning the fibres in dry-laid nonwovens, and also the presence of CMC on the fibres processed through the wet-laid method, which acted as impurities and prevented the homogeneous infiltration of CF during the further processing (Wölling et al., 2017).

Furthermore, it has been reported that the slivers of rCF/PET with the weight ratio of 60:40 developed through the carding process can be used to prepare the prepreg (Akonda et al., 2014). To this end, the slivers went through the gilling process to improve the fibres alignment and then prepreg tapes were prepared through assembling 10 slivers in parallel followed by heat treatment. Then, the non-crimp carbon fabrics were produced from the stabilised slivers after stitch-bonding using a PET filament. Analysing the mechanical characteristics of the prepared prepregs exhibited a lower performance compared with commercial

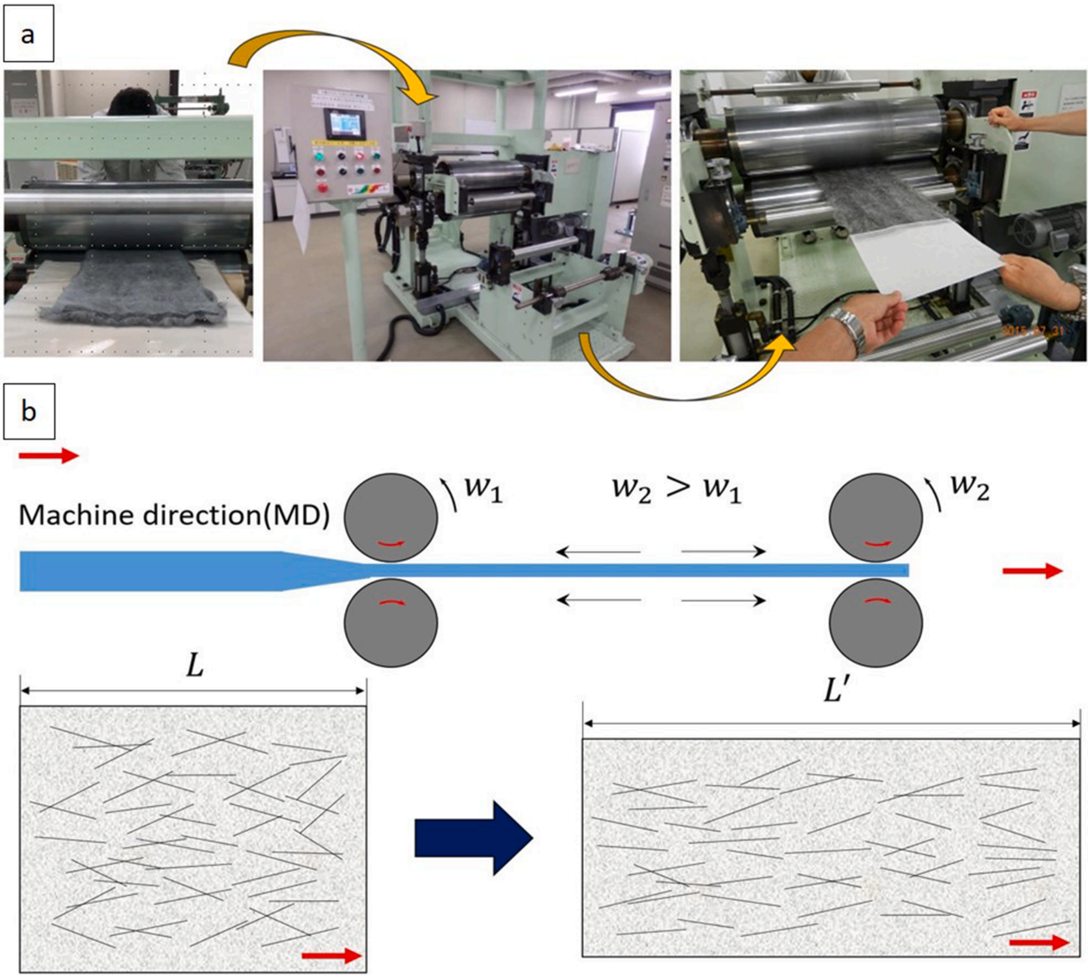

Fig. 15. a) Stretching and calendar pressing process for partially melting PA6 fibres for thermal bonding of CF/PA carding webs, b) impact of stretching process on the alignment of staple CF (Reprinted with permission from ref (Xiao et al., 2019), Copyright 2019 Elsevier).

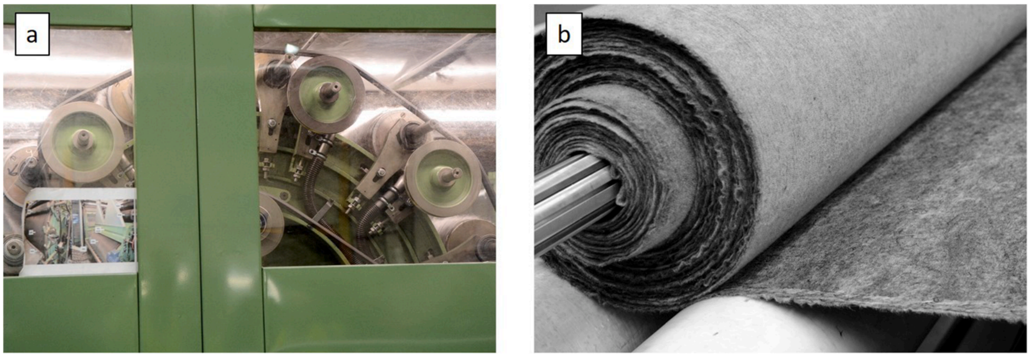

Fig. 16. a) The EGL Carbon Fibre company's carding machine, b) carbon fibre/thermoplastic nonwoven mat produced by EGL Carbon Fibre (Reprinted with permission from ref (Holmes, 2018), Copyright 2017 Elsevier).

CF/PET prepregs. The samples prepared from short recycled fibres had tensile strengths of 180.7 MPa and 260.5 MPa in both 0 and 0/90 layup directions and tensile modulus of 34.2 GPa. . The values for the commercial CF/PET 50:50 samples were 445.0 MPa and 38.0 GPa, respectively, indicating the superior performance of commercial prepregs (Akonda et al., 2014). Fig. 17 highlights the effective parameters on the final quality of nonwovens and yarns which should be adjusted for the processing the CF scrap.

5. Conclusion and future prospect

This article has reviewed different methods of recycling carbon fibre reinforced composites and also dry carbon fibre waste produced as offcuts during the manufacturing processes. Diverse aspects of three main recycling routes including mechanical, thermal and chemical methods and the influential parameters were discussed. The impacts of each recycling method on surface chemistry and mechanical properties of fibres were highlighted. Among these methods, the pyrolysis and mechanical processing demonstrate better potential to be used in

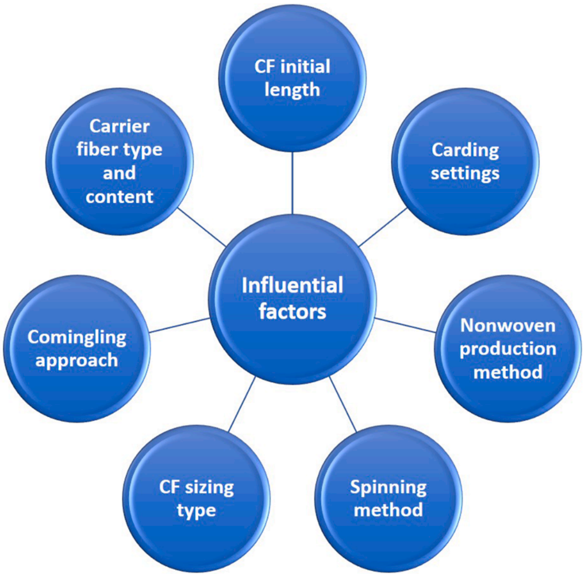

Fig. 17. Effective parameters on the processing of CF-scrap for producing hybrid CF yarns and nonwovens.

industry scale applications. However, researchers are still working towards ensuring the characteristics of recycled fibres are as close as possible to the virgin carbon fibre. In the main, there are obstacles in achieving large scale application of recycled carbon fibres in different fields. For instance, the low reproducibility of the recycled products is a challenge because there are currently no sorting mechanisms available based on the origin of the fibres used in the composite scraps. The lower mechanical characterises and altered surface chemistry of the recycled fibres are further issues that need to be addressed. All these problems have meant that the markets for recycled carbon fibre and composite materials have focussed upon non-structural applications. This prevents establishing a true cradle-to-cradle loop for CFRC within a circular economy context. The need to establish accurate and thorough modelling approaches to evaluate all the environmental and economic viabilities of these recycling methods is imperative if decision makers are to have comprehensive understanding on the applicability, cost, and environmental benefit of recycled components. Some areas such as using the recycled fibres in producing composites, tunning the interfacial interactions between fibres and matrix and optimising the recycling methods are recommended for further explorations. Also, developing procedures for obtaining value added products using recycled fibres and developed nonwovens and yarns for practical applications is another area to focus. Different developed methodologies for processing the dry carbon fibre waste was also reviewed. In the main, two main methods including the spinning of hybrid yarns and developing nonwoven products appear to have a promising future but improving the low mechanical performance of the developed products also remains a challenge.

The recycling of carbon-fibre based products is an emerging research area which will be playing a significant role in establishing circular economy approach for this high-tech fibre in the near future. Therefore, further research is required to improve the quality of fibres and lowering the environmental footprints of these methods.

Declaration of Competing Interest

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

Acknowledgement

We acknowledge support from the Australian Research Council World Class Future Fibre Industry Transformation Research Hub (IH140100018).

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