

Current status of recycling of fibre reinforced polymers: Review of technologies, reuse and resulting properties

Oliveux, Géraldine; Dandy, Luke O.; Leeke, Gary A.

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Current status of recycling of fibre reinforced polymers: Review of technologies, reuse and resulting properties



Géraldine Oliveux*, Luke O. Dandy, Gary A. Leeke

School of Chemical Engineering, University of Birmingham, Edgbaston, Birmingham B15 2TT, United Kingdom

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ABSTRACT

A complete review of the different techniques that have been developed to recycle fibre reinforced polymers is presented. The review also focuses on the reuse of valuable products recovered by different techniques, in particular the way that fibres have been reincorporated into new materials or applications and the main technological issues encountered. Recycled glass fibres can replace small amounts of virgin fibres in products but not at high enough concentrations to make their recycling economically and environmentally viable, if for example, thermolysis or solvolysis is used. Reclaimed carbon fibres from high-technology applications cannot be reincorporated in the same applications from which they were recovered, so new appropriate applications have to be developed in order to reuse the fibres. Materials incorporating recycled fibres exhibit specific mechanical properties because of the particular characteristics imparted by the fibres. The development of specific standards is therefore necessary, as well as efforts in the development of solutions that enable reusers to benefit from their reinforcement potential. The recovery and reuse of valuable products from resins are also considered, but also the development of recyclable thermoset resins. Finally, the economic and environmental aspects of recycling composite materials, based on Life Cycle Assessment, are discussed.

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* Corresponding author. Tel.: +44 121 414 5081.

E-mail addresses: g.oliveux@bham.ac.uk (G. Oliveux), LXD139@student.bham.ac.uk (L.O. Dandy), g.a.leeke@bham.ac.uk (G.A. Leeke).

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1. Introduction

Fibre reinforced resins, thermosets as well as thermoplastics, are increasingly used to replace metals in numbers of industrial, sporting and transport applications. One of the biggest challenges posed by fibre reinforced composites is their recycling. Environmental legislation is becoming more and more restrictive, and just the environmental impact of these materials disposed in landfills is accelerating the urgency to reach more industrial scale solutions to the recycling of composites. Landfill is a relatively cheap disposal route but is the least preferred waste management option under the European Union's Waste Framework Directive [1], and opposition to it is expected to increase over the coming years; it is already forbidden in Germany, and other EU countries are expected to follow this route [1]. Many different recycling techniques have been studied for the last two decades: mechanical processes (mainly grinding) [2–18], pyrolysis and other thermal processes [19–39], and solvolysis [40–85]. Some of them, particularly pyrolysis, have even reached an industrial scale, and are commercially exploited: for example, ELG Carbon Fibre Ltd. (ELGCF) in United Kingdom use pyrolysis [32], Adherent Technologies Inc. (ATI) in USA use a wet chemical breakdown of composite matrix resins to recover fibrous reinforcements [81] and, in France, Innoveox [82,83] propose a technology based on supercritical hydrolysis. Pyrolysis is the most widespread technology as it is a proven and heavily used process in the chemical industry. However as the fibres degrade at high temperatures, solvolytic processes have attracted increasing interest, especially over the last decade. Supercritical fluids have received much attention because of their tuneable properties depending on operating conditions (temperature, pressure and volume), however the associated equipment can be very expensive due to the severity of the conditions. Recent investigations have considered less severe conditions, but in detriment to process time [53,56,76,78,79]. Solvents and/or catalysts are used that can be toxic and difficult to dispose or separate. The fibres can also be more damaged by the use of catalysts. If the objective of recycling is to recover fibres, this cannot be done in detriment of environmental aspects (used energy, chemical products, emissions. . .). A complete evaluation must be carried out in order to compare the different technologies in terms of environmental impact, efficiency and commercial viability. It is also quite clear that the choice of the separation/recycling

method depends on the material to recycle and on the reuse applications of the fibres in particular. The recovered products, mainly fibres and products from resin decomposition, are most often systematically characterised and show that they can be reused. In particular recycled carbon fibres (rCFs) (often discontinuous as the materials require to be cut in smaller pieces before treatment) have been incorporated with success in a few trials [2,16–18,72,76,86–98]. Recycled glass fibres (rGFs) can also be reused as fillers in thermoplastics or short fibres in BMC, mainly [3,4,6,7,13]. On the other hand, fractions containing products from resin degradation by solvolysis have received very little consideration. Pyrolysis products from the resin have been mainly considered as a source of energy to feed back into the process.

Currently solutions do exist to recycle composite materials. It can be seen in the literature that many different processes and methods have been applied and have shown the feasibility of recycling such materials, some of them being more commercially mature than others. However, industrial applications using recycled fibres or resins are still rare, partly because of a lack of confidence in performance of rCFs, which are considered as of lower quality than virgin carbon fibres (vCFs) [99], but also because rCFs are not completely controlled in terms of length, length distribution, surface quality (adhesion to a new matrix) or origin (often different grades of fibres are found in a batch of recycled composites coming from different manufacturers). Furthermore, recycling of composites is globally not closed-loop in terms of resource efficiency as recycled fibres cannot be reused in the same applications as their origin. For example if they come from an aircraft structural part, they cannot be reused in a similar part. In light of this, relevant applications have to be developed and specific standards are required in order to manage those reclaimed fibres. Demonstrators have been manufactured with rCFs, showing potential new applications [34].

The article presents a review of technologies that have been investigated for about 20 years to recycle composite materials, from industrially exploited processes to methods still in research. The review covers applications for both reclaimed fibres and products recovered from degraded resins, as well as some economic and Life Cycle Assessment (LCA) aspects.

2. Review of recycling technologies and discussion

Recent articles have updated the state of the art of the recycling of composite materials [34,83,100,101], but they are orientated towards carbon fibre reinforced composites (CFRCs) and near- and supercritical solvolysis [83], whereas the vast majority of composites being recycled (about 98% in weight [102]) concerns glass fibre reinforced composites (GFRCs). CFRCs show a high commercial value due to the presence of carbon fibres, which is probably the reason why they benefit from more attention.

A review of the different existing methods applied to recycle composites is presented and discussed, and highlighted according to the materials to recycle and the potential reuse applications.

2.1. Mechanical recycling

This technique consists of grinding materials more finely after a first crushing or shredding step into smaller pieces [2]; the latter is common to all the recycling techniques. Generally different sizes of recyclates can be recovered and separated by sieving into resin-rich powders (and rich in fillers for SMC materials for example) and fibres of various lengths that are still embedded in resin. Flakes of materials can also be found in the recyclates. Mechanical grinding has been more applied to glass fibre reinforced composites [3–13], in particular SMC and BMC [13], but work on CFRCs also exists [2,14–18]. One of the first evaluations of the potential of this technique was published in 1995 by the Clean Washington Center where Kevlar[®] aramid and carbon fibre reinforced scrap edge trimmings from moulded composites and cured prepreg composites were considered [2]. The use of ground composite materials can have two purposes: filler or reinforcement. Their use as filler is not commercially viable because of the very low cost of virgin fillers such as calcium carbonate or silica, this is particular pertinent if CFRCs are considered. The fillers concerned are the powdered products recovered after sorting. The incorporation level of filler material is quite limited (less than 10 wt.%) because of the

deterioration in mechanical properties and increased processing problems at higher contents (due to higher viscosity of the compound) [13]. If they cannot competitively replace currently used fillers, they could be used as an energy source as they are rich in resin. CF reinforced polyether-ether-ketone (PEEK) resin was studied by Schinner et al. [14]. The authors observed that cutting mills gave more homogeneous fibre length distribution and longer fibres than hammer mills although the cutting blades wore faster. The ground materials were then successfully incorporated into a virgin PEEK resin and moulded by injection or press up to 50 wt.%. A direct reforming process without grinding was also successfully performed, however this was not applied to end-of-life materials. The fibrous fractions of ground thermoset materials are reported to be more difficult to reuse, in particular as reinforcement and, even with low reincorporation, the resulting mechanical properties are significantly impaired [13] due to a poor bonding between the recyclates and the new resin. However Palmer et al. [4] showed that when the fibrous fractions (from glass fibre SMC) similar to virgin fibre bundles were incorporated at about 10 wt.% in DMC, the mixing time can affect positively the mechanical properties of the resulting materials. A longer mixing time of the paste with the recyclate enabled improved mechanical properties compared to a standard mixing time and furthermore properties comparable to the standard material could be achieved. This is explained by an improved interface between the recyclate and the new resin.

Industrial applications of this technique actually exist largely among glass fibre reinforced composite (GFRC) manufacturers, like Mixt Composites Recyclables (M-C-R), a subsidiary of Plastic Omnium in France, Filon Products Ltd. in United Kingdom and a few others as shown in Table 1. There is no known grinding process exploited industrially to treat CFRCs. Other processes which allow a separation of the fibres and the matrix (like the ones presented here after) are preferred for carbon fibres as they can be recovered without contamination and potentially reused as reinforcement in new composites.

Recently a novel grinding process was proposed by Roux et al. [103] that used electrodynamic fragmentation to shred carbon fibre reinforced thermoplastic. In this method the material is placed in water between two electrodes, and a high voltage, between 50 and 200 kV, is then applied to fragment the material into smaller pieces.

Table 1

Companies using mechanical grinding as recycling technology; grey cells indicate companies no longer active and white cells for companies still active.

Company	Country	Comments/product range
Mixt Composites Recyclables (MCR) – group Plactic Omnium [5]	France	Micronised powder that can be used in SMC and BMC manufacturing Ground GFs RECY-FIB® that can be used as reinforcement in asphalt, concrete, chipboards, composite materials
Filon Products Ltd. [6]	UK	Ground GFRCs reincorporated in their products
Reprocover [7]	Belgium	Different types of cover (access chamber, inspection chamber,...), street furniture (ashtray, litter bin, letter box...) and more recently bi-block railway sleeper
Hambleside Danelow [8]	UK	Ground GFRCs used within their main stream production of GFRC or injection moulded polypropylene
Phoenix Fibreglass Inc. [9]	Canada	Chopped strand GF (about 0.5 in. length) and milled GF (less than 0.5 in. length) that can be used as partial reinforcement in new BMC
ERCOM [10]	Germany	Coarse and fine fillers that can replace virgin fillers like calcium carbonate 3 powder grades depending on the size of the particles and 3 fibrous grades depending on the average length of the fibres
Fiberline – Zajons – Holcim AG/Geocycle [11]	Denmark and Germany	GFRC waste from Fiberline ground by Zajons and used for energy recovery in cement kiln
Eco-Wolf, Inc. [12]	Florida, USA	Global system composed of Eco-Grinder™ (grinding of GFRC waste while maintaining fibre integrity) and Eco-Dispensing Macerator™ (meters and transports the ground materials to existing spray-up equipment) Ground composites can produce fibres from 1 in. to a fine powder FRP Green is a distributor of the global system in Oceania

2.2. Thermal processes: pyrolysis, fluidised bed pyrolysis, micro-waves assisted pyrolysis

Thermal processes include pyrolysis, fluidised-bed pyrolysis and pyrolysis assisted with micro-waves [19–31]. These techniques allow the recovery of fibres, eventually fillers and inserts, but not always the recovery of valuable products from the resin (i.e. monomers that could be reused to produce resins). The resin is volatilised into lower-weight molecules and produces mainly gases such as carbon dioxide, hydrogen and methane for example, and an oil fraction, but also char on the fibres. The processes operate between 450 °C and 700 °C depending on the resin. The lower temperatures are adapted to polyester resins, whereas epoxides or thermoplastics, like PEEK for example, require higher temperatures.

Even higher temperature processes are also possible but concern combustion with energy recovery found in cement kilns, in which the composite waste is converted into energy and into raw materials components for the cement (fillers and fibres). However no more than 10% of the fuel input to a cement kiln could be substituted with polymer composites reinforced with glass fibres. Indeed the presence of boron in E-glass fibres (the most widespread) was found to affect the performance of the cement [13]. However Jean-Pierre Degré, Senior Vice President, Holcim Group Support, Sustainable Development – Alternative Resources, confirmed that co-processing of glass fibre reinforced composites waste as alternative fuel at Lagerdorf has no negative effect on the quality of the cement produced [104], however the incorporation proportions are not given. According to the European Plastics Converters (EuPC), the European Composites Industry Association (EuCIA) and the European Recycling Service Company (ECRC), “the European composites industry considers the cement kiln route to be the most sustainable solution for waste management of glass fibre reinforced thermoset parts” [105]. However at present this solution is not economic compared to landfill where landfill is an option [104].

2.2.1. Pyrolysis

The most studied thermal process is pyrolysis performed in absence or in presence of oxygen, and even more recently in presence of steam [31]. The matrix degradation produces an oil, gases and solid products (fibres, eventually fillers and char). The fibres are contaminated by this char and require a post-treatment in a furnace at 450 °C at least to burn it, for example for GFRC [21] (Fig. 1). This also leads to a higher degradation of the fibres. This process has been more developed to recycle carbon fibre reinforced matrices and has reached commercially exploited industrial scale, as shown in Table 2.

Glass fibres suffer from the high temperatures and their mechanical properties are decreased by at least 50%, especially as the minimal process temperature is 450 °C [21]. Carbon fibres are less sensitive to temperature but they can be contaminated by a char-like substance remaining from the degradation of the resin, which prevents a good bond with a new resin. At 1300 °C this substance is completely removed and the fibres are perfectly clean with highly activated surface, but their strength is significantly reduced [24]. A small number of mechanical property values measured by different work groups were gathered by Pimenta and Pinho [34]. Additional values were also found and are reported in Table 3. They show that the tensile strength can be reduced by up to 85%, but can also be unaffected by the treatment. The treatment conditions thus play a great role on the resulting fibre properties. A lower reduction in tensile strength was observed when fibres were reclaimed from a composite than when they were heated in air on their own (so not embedded in a resin) [13]. Above 600 °C the tensile strength of the rCF was reduced by over 30%. The fibres also seem to have different sensitivity to pyrolysis conditions depending on their type [24,25,90]. For example, Hexcel AS4 carbon fibres showed a strong oxidation from 550 °C in oxygen, whereas Toho-Tenax high tenacity carbon fibres (HTA) were not oxidised below 600 °C in air (Fig. 2a and b respectively). Air is less oxidant than neat oxygen. In oxidant conditions epoxy resins are more easily degraded than in inert conditions and at temperatures in the range of 500–600 °C it is possible to completely remove resin residues [24]. A compromise is thus necessary between resulting mechanical properties and the amount of remaining resin residue. HTA fibres recovered after a first step at 550 °C in nitrogen during 2 h and a second step at 550 °C in oxidant conditions retained more than 95% of their tensile strength without resin residue on the surface [24].

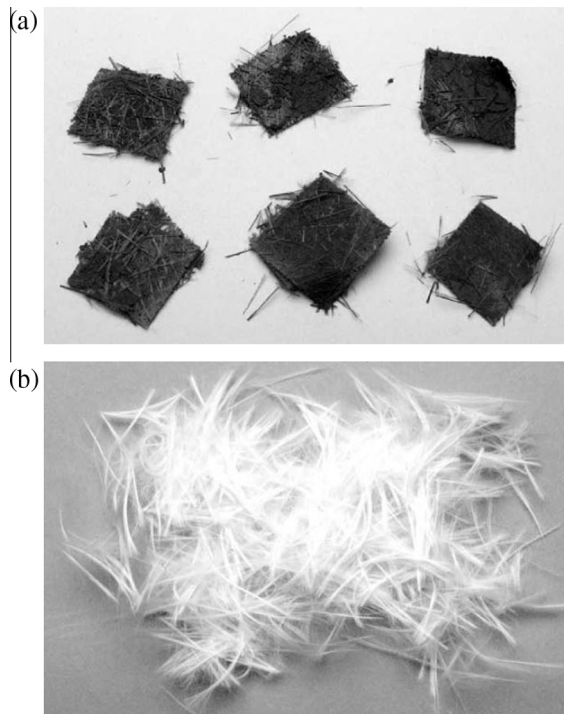


Fig. 1. (a) Solid pyrolysis residues, (b) recovered fibre after separation [21].

A pyrolysis temperature in the range of 500–550 °C appears then to be the high limit of the process in order to maintain acceptable strength for carbon fibres; whereas glass fibres retain less than 50% of their mechanical properties at the minimal temperature of 400 °C [13]. This type of technique appears therefore to be more adapted to recover carbon fibres. Finally, in a real industrial process recycled carbon fibres are reclaimed from a diverse feedstock based on different types of carbon fibres with varying properties. They are mixed together during the separation process so that it becomes hard to compare properties of single fibres to those of virgin fibres. Fibres reclaimed from industrial processes ultimately present a distribution of properties. As explained by ELGCF [32], they blend their products in order to minimise property variation. They claim that the fibre properties are approximately 90% of those of virgin fibres following their pyrolysis process. It was reported that laboratory-scale or pilot-plant pyrolysis led to better results in terms of fibre surface quality and mechanical properties than the ones obtained with industrial-scale processes [95].

2.2.2. Fluidised-bed process

Fluidised-bed process has been applied to the recycling of glass fibre reinforced composites (GFRCs) [23,26] and CFRCs [13]. This pyrolysis-based process uses a bed, of silica sand for example, fluidised by hot air so conditions are oxidant. It enables a rapid heating of the materials and release the fibres by attrition of the resin. As in classical pyrolysis, a small amount of oxygen is required to minimise char formation. A rotating sieve separator was implemented in Pickering's process to separate fibres from fillers of recycled GFRCs [23]. The organic fraction of the resin was further degraded in a secondary combustion chamber at about 1000 °C, producing a clean flue gas (for energy recovery). At 450 °C glass fibre tensile strength was reduced by 50%, while at 550 °C the reduction achieved 80%. Carbon fibres show a lower strength degradation of about 25% when processed at 550 °C [55]. Analysis of their surface showed that the oxygen content resulted in a little reduction, indicating that the fibres have good potential for bonding to a polymer matrix [13]. The interest of this process is that

Table 2

Companies using pyrolysis as recycling technologies; green cells indicate pilot plant, white cells for industrial or semi-industrial plant and grey for no longer active companies.

Company	Country	Comments/product range
ELG Carbon Fibre [32]	UK	Unsize milled fibres (average lengths 100 and 200 μm) or granules containing 100 μm -length fibres Unsize chopped fibres (random) with lengths between 6 and 60 mm Prices: boxes = 18 €/kg, pallets = 13 €/kg and tonnes = 11 €/kg
Materials Innovation Technologies RCF (MIT-RCF) [34]	USA	Fibre reclamation as well as development of manufacturing processes (3-DEP) Preforms and finished parts
Karborek Spa [35]	Italy	Milled CFs in pellets Chopped CFs Multi-layered felt containing 95% of recycled fibres
CFK Valley Stade Recycling GmbH (Karl Meyer Group) [36]	Germany	Products distributed by the company CarboNXT GmbH within the Karl Meyer group Chopped and milled fibres
Hadeg Recycling Ltd. [37]	Germany	Mean fibre length of 200 μm Other types of fibres possible
ReFiber ApS [24]	Denmark	No longer active
Japan Carbon Fiber Manufacturers Association (JCMA) [34]	Japan	Pyrolysis and grinding
Firebird Advanced Materials [38]	USA	Microwave-assisted pyrolysis No longer active
Formoso Technologies Group [39]	Spain	A pilot plant developed to treat waste of production (GF fabric, carbon fibre rolls, non-conform uncured prepregs and non-conform cured parts) Recovery of oil for energy supply from the resins and of fibres

it can treat mixed and contaminated materials, with painted surfaces or foam cores in composites of sandwich construction or metal inserts [13]. This process is therefore particularly suitable for end-of-life waste, however it has not largely been applied to reclaim fibres, in particular carbon fibres. Furthermore the fluidised bed process does not allow recovery of products from the resin apart from gases, whereas pyrolysis can enable the recovery of oil containing potential valuable products. Carbon fibres seem to be more damaged than with pyrolysis; however the process has not been optimised. In addition to the high temperature, attrition by the fluidised sand might also damage the fibres.

2.2.3. Micro-wave assisted pyrolysis

Microwave-assisted pyrolysis has also been considered over the last ten years to recover carbon and glass fibres by Lester et al. at the University of Nottingham [27], by the American company Eltron Research [33], and more recently by Åkesson et al. at the University of Borås [28]. The main advantage of microwaves is that the material is heated in its core so that thermal transfer is very fast, enabling energy savings. Microwave-assisted pyrolysis heats composite wastes in an inert atmosphere, degrading the matrix into gases and oil. The first application of this heating method to recycle composites was studied by Lester et al. [27] in 2004. They used quartz sand to suspend the samples made of carbon fibres and epoxy in a microwave cavity and glass wool to prevent solids leaving the cavity. Microwave treatment was also used by the American company Firebird Advanced Materials at the same time, but they stopped their activity due to a lack of investors in 2011.

In Sweden the project involving the University of Borås and Stena Metall Group used this method to recycle wind turbine blades made of glass fibres and a thermoset resin (apparently polyester according to the nature of the products recovered from the resin [28]). Three kg of ground materials were pyrolysed in a 10 L reactor at 440 °C during 90 min and an oil and glass fibres were recovered and analysed. A non-woven mat was then manufactured with the recycled fibres and used in a new material by alternating these new mats with virgin glass fibre (vGF) mats. However as the recovered fibres were coated by residual char, the adhesion with a new matrix was unsatisfactory, which led to poor

Table 3

Single fibre testing results obtained on fibres recovered by pyrolysis (r = recycled and v = virgin).

	Process	Fibre type	Single fibre tensile strength			
Kennerley et al. [106]	Fluidised bed pyrolysis	Owens Corning GF roving RO7E for SMC	v	10 mm	2.61	GPa
			r	10 mm	–52% –64%	at 450 °C at 550 °C
Hyde et al. [55]	Comparison between different processes	Grafil 34-700	v		4.09	GPa
				Fluidised bed Microwave scPrOH	–25% –20% –5%	
Meyer and Schulte[24]	ReFiber optimised pyrolysis	Toho-Tenax HTA	v		3.712	GPa
			r		–4%	
Akonda et al. [93]	ELGCF pyrolysis	TR50S CF from Pyrofil	v	25 mm	3.19	GPa
			r	25 mm	–10%	at 500 °C – 10 min in air
Pimenta 2012 [107]	ELG pyrolysis	Hexcel AS4	v	10 mm	4.897	GPa
				20 mm	4.551	GPa
			r-A	10 mm	–83%	Decreasing temperature ↓
				20 mm	–85%	
			r-B	10 mm	–75%	
				20 mm	–81%	
			r-C	10 mm	–67%	
				20 mm	–76%	
r-D	10 mm	–2%				
	20 mm	–4%				
Greco et al. [108]	ENEA pyrolysis process used by Kaborek Spa	Torayca T800S	v		5.9	GPa
			r	10 mm	–17%	
				20 mm	–24%	
			r 450	10 mm	–17%	Post-treated at 450 °C
				20 mm	–36%	
			r 600	10 mm	–69%	Post-treated at 600 °C
				20 mm	–82%	
			r acid	10 mm	–24%	Post-treated in nitric acid
	20 mm	–44%				
Stoeffler et al. [96]	Materials Innovation Technologies pyrolysis	Carbon fibres (diverse feedstock)	Loss of 22% compared to virgin T700S fibres from Toray			

mechanical properties when only recycled fibres were in the composites. The amount of recovered fibres has to be limited to 25 wt.% in order to obtain acceptable mechanical properties but they were not as high as the equivalent material made of vGFs (a decrease of about 25% was observed). The quality of the new mat also affected the mechanical properties of the overall composite as the fibre length distribution was quite wide (about 3–30 mm, Fig. 2 in [28]) and the fibres were heterogeneously distributed in the mat.

2.3. Solvolysis

2.3.1. Principle

Solvolysis consists of a chemical treatment using a solvent to degrade the resin. This technique was first considered about 30 years ago [40] and applied to unsaturated polyesters (UP) and SMCs as UP is

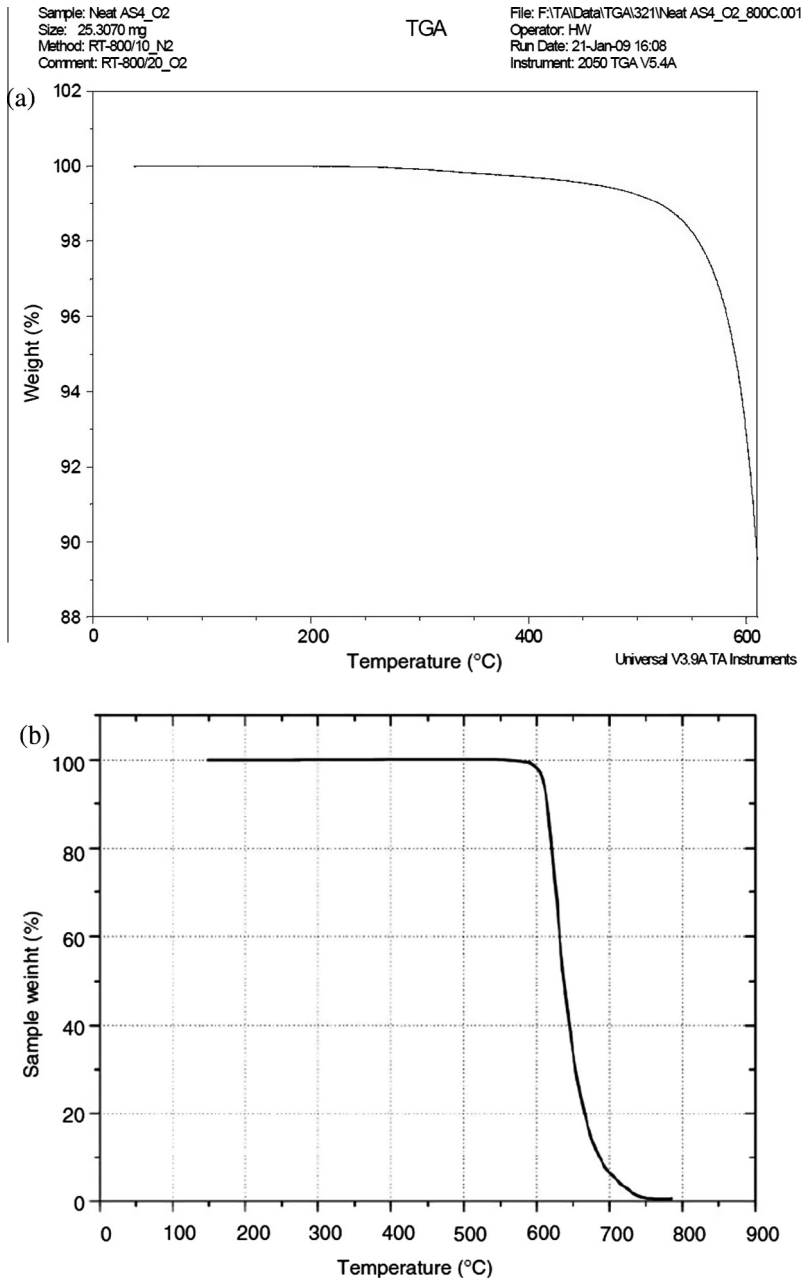


Fig. 2. (a) TGA of AS4 carbon fibres in oxygen [25], (b) TGA of HTA carbon fibres in synthetic air [24].

one of the most widely used thermoset resins and in particular in SMCs. Hydrolysis between 220 and 275 °C either with or without added solvent or catalyst was used by Kinstle et al. in the 1980s to degrade UP into its monomers (carboxylic acids and glycols) and a styrene–fumaric acid copolymer [40]. Since then many different conditions and solvents were tried in order to recycle thermoplastics, thermosets and their fibre reinforced composites [40–85]. Solvolysis offers a large number of

possibilities thanks to a wide range of solvents, temperature, pressure and catalysts. Its advantage, compared to pyrolysis, is that lower temperatures are generally necessary to degrade the polymers, in particular UP and epoxides. However when supercritical conditions, of water for example, are reached, reactors can become expensive as they have to withstand high temperatures and pressures, as well as corrosion due to modified properties of the solvents [109]. A reactive solvent, sometimes in mixture with a co-solvent or with a co-reactive solvent, diffuses into the composite and breaks specific bonds. It is therefore possible to recover monomers from the resin and to avoid the formation of char residues. Depending on the nature of the resin, more or less high temperatures and pressures are necessary to degrade the resin. Polyester resins are generally easier to solvolyse than epoxy resins and so require lower temperatures to be degraded. During the last decade this method has been more intensively used to recycle composites, in particular CFRP, as the recovery of carbon fibres has become a commercial interest.

Among all the tested solvents, water appears as the most used, sometimes neat [40,42,47,49–51,54,62,71,72], and sometimes with a co-solvent (alcoholic, phenolic, amine) [40,53,59,71]. Often it is used with alkaline catalysts like sodium hydroxide (NaOH) or potassium hydroxide (KOH) [42,51,52,62,66,72,75,84], but less often with acidic catalysts [44,51,53,65,76]. Acidic catalysts were mainly used to degrade more resistant resins, for example PEEK, or to degrade epoxy resins at low temperatures. A few other solvents have also been used, mainly alcohols like methanol, ethanol, propanol, and acetone or even glycols, with or without additives/catalysts [40,43,52,55–57,61,62,64,69,70,73,77–79,85].

Numerous lab-scale experiments have been carried out, but only a few studies have reached industrial or semi-industrial scale (Table 4). ATI and more recently Innoveox have proposed to sell or licence their technology (Table 4). In addition Panasonic Electric Works show a willingness to exploit their hydrolysis process to recycle 200 tons of GFRC manufacturing wastes annually [66].

Depending on the amount of solvent and on temperature, the fluid can be vapour, liquid, biphasic or supercritical. When the fluid is in vapour phase or has a gas-like density in the supercritical fluid state, the process is more a thermal process than a solvolysis (e.g., hydrothermal processes using water as solvent – see [84] for example).

2.3.2. High temperatures (>200 °C) and pressures (HTP)

Since 2000, supercritical conditions have gained more attention due to the tuneable solvent properties which significantly change from subcritical to supercritical conditions. Supercritical fluids (SCFs) show properties intermediate between liquid and gas phases. They have low viscosities, high mass transport coefficients, high diffusivities, and a pressure dependent solvent power [55]. The involved chemistry is affected by these changing solvent properties. This offers the capability of controlling

Table 4

Companies using solvolysis as recycling technologies; green cells indicate pilot plant, white cells for industrial or semi-industrial plant.

Company	Country	Technology	Comments/product range
Adherent Technologies [58]	USA	3-step process combining pyrolysis and solvolysis	Only propose to sell or license the technology, providing support on building and operating a facility A rough evaluation gives a cost of about \$4 Million for a 2 tonne/day stand-alone facility
Panasonic Electric Works Co. [66]	Japan	Hydrolysis	Monomers of the resin (mainly glycols) and a styrene–fumaric acid copolymer (SFC) which could be used as a low profile additive. The solid fractions (fibres and fillers) are suggested to be used as filler in new materials
SACMO [72]	France	Solvolyysis	The company does not exploit the technology but designed and manufactured a reactor prototype
Siemens AG [80]	Germany	Solvolyysis	The technology has been registered for a patent
Innoveox [82]	France	Hydrothermal Oxidation	Only propose to sell their technology with their support

the solvent properties and reaction rates and selectivities through pressure manipulations [111]. Water in particular has been considered because of its temperature and pressure dependent properties. Depending on the conditions, it can support ionic, polar non-ionic or free-radical reactions, so that it is said to be an adjustable solvent [110]. Supercritical water (SCW) (temperature >374 °C and pressure >221 bar) has been mainly applied to CFRC in order to recover carbon fibres of good quality without paying much attention to the products of the resin degradation. This is because of the potential high commercial value of rCF; however the intense hydrolysis conditions require specific and expensive reactors. Alternative solvents with lower critical temperature and pressure have been considered, mainly ethanol, methanol, propanol and acetone, as well as additives or catalysts added to water in order to moderate the operating conditions. Supercritical alcohols or acetone, for example, actually require temperatures as high as pure water in order to achieve a sufficient elimination of resin from the carbon fibres [62,63], however the pressure is much lower than with water (about 255 bar at 450 °C for 1-propanol compared to about 630 bar with the same amount of water). Only a catalyst and semi-continuous conditions enabled a significant reduction in the temperature [63]. The catalyst appears to modify the reaction pathway [112]. It appears furthermore that efficient temperature and pressure levels depend deeply on the type of epoxy resin [63,65,72,73] and more widely on the type of resin. Indeed Elghazzaoui [72] observed that RTM6 epoxy resin requires supercritical conditions of water to be degraded, whereas subcritical conditions at about 350 °C were sufficient to degrade the 914 epoxy resin, both from Hexcel. A bisphenol-A (BPA) epoxide cured with 1,2-cyclohexane dicarboxylic anhydride could be completely eliminated from carbon fibres in supercritical methanol at 350 °C [73], whereas LTM26EL epoxy resin from Cytec, which contains cresol and BPA cured with amine agents, requires about 450 °C to be eliminated [63]. In light of this, it is hard to compare the results obtained at different conditions when the resins are different. Regardless of epoxy resin type and conditions, phenol was the main degradation product. Other phenolic compounds were identified such as isopropyl phenol and cresol depending on the conditions and the epoxy type, as well as anilines when the curing agent was an amine. Gases were also produced but were not always mentioned nor analysed. Elghazzaoui [72] identified hydrogen, oxygen, carbon dioxide, methane, ethane and propane in particular from the degradation of the 914 epoxy resin from Hexcel. Thermoplastic resins like PEEK appear to be harder to degrade due to their high thermal stability. It is generally necessary to reach at least their melting temperature, which is about 345 °C for PEEK. Until now only supercritical water was reported to be efficient to degrade PEEK, producing mainly phenol as the degradation product [51,52]. Otherwise strongly acidic or alkaline conditions are necessary.

For UP, lower temperatures and pressures are required, generally below 300 °C [40,41,54,60,69,71,109]. Mainly water was used to degrade UP, as hydrolysis is the reverse reaction of esterification. Mixtures with alcohols and amines were also tried [54,60] or with ketones [40–42], as well as neat alcohols [63,71]. The addition of a catalyst like sodium hydroxide (NaOH) did not show a significant effect to lower conditions and was extremely damaging for the glass fibres above 300 °C [71]. Mixtures with alcohols and amines showed a significant effect on the process yield, which was explained by transesterification occurring in presence of alcohols [60]. In most cases only the resin degradation was studied, GF were almost never considered except in reference [71]. This is due to the low commercial value of GF and to their fragility when they are exposed to thermal, acidic and alkaline conditions.

At small scale, the reaction rate is not limited by diffusion [110], however at larger scale (20 L), diffusion limiting effects due to the scale up and to the selection of larger pieces of composites could be observed as shown in Fig. 3. For this reason SCFs have been used as they behave like gases, showing low viscosities, high mass transport coefficient and diffusivity and a pressure dependent solvent power [55]. It should also be noted that if the resin concentration is too high, the liquid medium becomes saturated and the reaction is slowed down [63,72]. Semi-continuous conditions were then used by Pintero-Hernanz et al. [63] to improve those aspects. This enhanced the diffusion processes; it may also avoid the deposit of resin residue on the fibres and the degradation of valuable products released from the resin.

After solvolysis in a batch reactor the recovered fibres are coated with an organic residue that requires a long and expensive rinsing to remove [71,72]. The characterisation of fibre surface after semi-continuous solvolysis in supercritical propanol showed that the oxygen content was much lower



Fig. 3. Evidence of diffusion limitation on a 50 × 50 mm GFRC sample after hydrolysis in a 20 L reactor (temperature assumed to be around 300 °C according to the report) [113].

for rCF than for vCF, which led to lower interfacial shear strength (IFSS) [114,115]. In oxidant conditions, it was possible to completely remove the resin and to leave no residue on the fibre surfaces, which then have an oxygen content higher than virgin fibres [67], however the tensile strength significantly decreased.

2.3.3. Low temperatures (<200 °C) and pressures (LTP)

LTP solvolysis is generally carried out below 200 °C and at atmospheric pressure. Catalysts and additives are necessary in order to degrade the resin as the temperature is very low, stirring can also be necessary [48,56,70,76,78,79]. Acid medium has been mostly used (nitric, sulphuric and acetic acids – [48,76,78] respectively) in comparison to HTP where alkaline conditions have been tested most often. Some acid solutions are very strong and can be very dangerous in terms of safety (health and environment). The only advantage of this method is that it offers a better control of the occurring reactions, and as the temperature is low, secondary reactions do not seem to occur. This enables a higher recovery of epoxy monomers, but not necessarily molecules of curing agent.

As shown in Table 5, the effects of LTP solvolysis on fibre properties are comparable to the ones observed at HTP, due to the use of strong acid or oxidant conditions. Finally, LTP methods use solutions that can be difficult to dispose of or to recycle.

2.4. Global discussion

Pyrolysis and solvolysis are the most preferred techniques to recycle composites, in particular CFRCs, both having the objective to reclaim fibres. Both techniques also require a first step of shredding or crushing into smaller pieces because of the size of some industrial parts in relation to the size of reactors. They both have proven to enable the recovery of carbon fibres largely maintaining their reinforcement capability, whereas glass fibres are quite damaged (Tables 3 and 5). Compared to pyrolysis, solvolysis is able to avoid the formation of char that contaminates the fibre surface and prevents good interaction between rCFs and a new matrix [58]. Semi-continuous conditions seem to be necessary to reclaim clean fibres without using oxidant conditions, otherwise a post-rinsing step is required to remove the organic residue that deposits during the cooling phase in batch reactors. Fibres recovered by solvolysis can be cleaner than those recovered by pyrolysis but that depends on the conditions employed (oxidant, very acidic or alkaline). When the fibres are woven in the composite material, it seems harder to remove the resin in particular between two intersecting tows by pyrolysis [95]. It would be pertinent to compare the efficiency of a solvolysis process to that of pyrolysis for the removal of residual resin as a result of improved mass transfer.

Table 5

Single fibre testing results obtained on fibres recovered by solvolysis (r = recycled and v = virgin).

	Process	Fibre type	Single fibre tensile strength			
Adherent Technologies 2006 [58]	Solvolysis	Carbon fibres	Loss <10% compared to virgin fibres			
Pinero-Hernanz et al. [62]	Supercritical water	Toray T600	v	up to:	4.09	GPa
			r		-11%	(up to)
Pinero-Hernanz [63]	Sub- and supercritical alcohols and acetone	Grafil 34-700	Loss of 1–15% compared to virgin fibres			
Yuyan et al. [65]	Subcritical water (<300 °C)	Carbon fibres	Loss of 1.8–4.1% compared to virgin fibres with or without acidic catalyst			
Bai et al. [67]	Supercritical water + O ₂	Toray T300	v	30 mm	3.11	GPa
			r	30 mm	0% –38%	at 96.5% purity at 100% purity
Okajima et al. [68]	Liquid subcritical water	Carbon fibres	Loss of 12% without catalyst Loss of 17% with K ₂ CO ₃ as a catalyst			
Okajima et al. [73]	Supercritical methanol	Carbon fibres	Loss of 9% compared to virgin fibres			
Sakuma et al. [70]	Subcritical benzyl alcohol + K ₃ PO ₄	Toray T300B	Tensile strength fully retained			
Liu et al. [74]	Subcritical water	20 mm	(315 °C) + phenol + KOH			
			v		GPa	Carbon fibres
Knight et al. [75]	Supercritical water + KOH	IM7	v	25 mm	5.25	GPa
			r	25 mm	+7%	
Elghazzaoui [72]	Subcritical water (350 °C)	Toray T300	Retention between 90% and 100% of the tensile strength			
Li et al. [78]	Pre-treatment in acetic acid + acetone/H ₂ O ₂	Toray T700	v		2.81	GPa
			r		-5%	at 80 °C
					-7%	at 100 °C
Oliveux et al. [71]	Subcritical water	E-glass Advantex®	v	10 mm	2.1	GPa
			r	10 mm	-67%	at 350 °C –5 min
					-52%	at 300 °C –30 min
					-33%	at 275 °C –120 min

The purity of recycled fibre surface has an effect on its capability to adhere to a new resin. When resin residues remain at the surface after solvolysis or pyrolysis, the single fibre tensile strength seems to be improved [67,71,107]. However, when reincorporated into a new composite, this residue reduces good interaction with the new matrix, leading to poorer mechanical properties. A study realised at the North Carolina State University compared rCFs recovered from end-of-life F18 stabiliser components by Milled Carbon's pyrolysis process (now ELGCF) and by ATI's process [88]. The analysis of the fibre surface showed that fibres recovered from pyrolysis and from ATI's process are comparable to each other in terms of oxygen content. While fibres recovered after the ATI's LTP process showed that some catalyst molecules remained on the fibre surface resulting in a poor resin to fibre adhesion, and consequently in poor mechanical properties when incorporated in an injection moulded polycarbonate resin [90]. ELGCF improved its pyrolysis process and produced rCFs clean enough to enable good adhesion with a new matrix, as can be seen in Fig. 4.

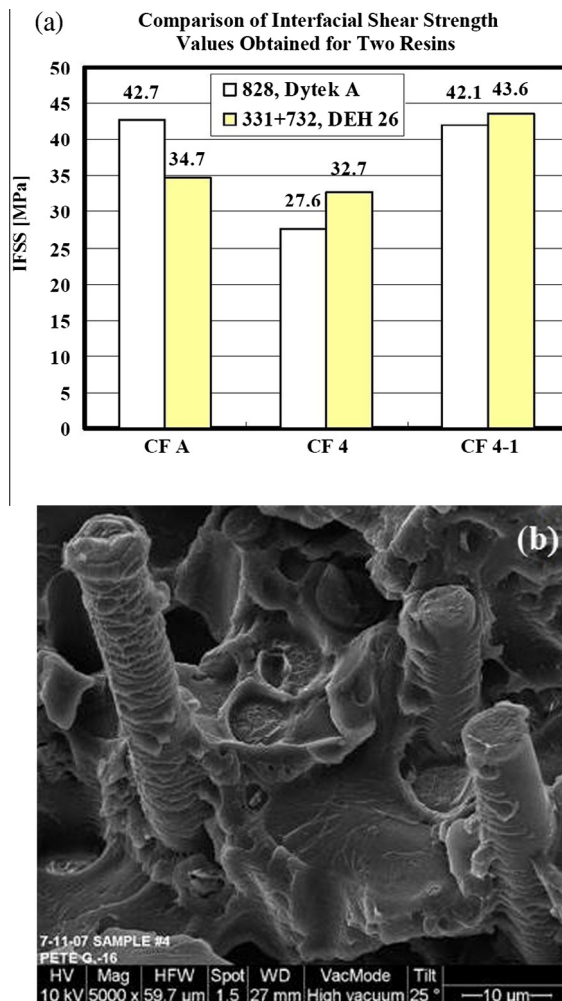


Fig. 4. (a) Measurements of resin to fibre adhesion with CFs reclaimed from ELGCF pyrolysis. CF A are unsized AS4 fibres, CF 4 are rCFs and CF 4-1 are rCFs from the optimised pyrolysis [86], (b) rCFs incorporated into a polycarbonate matrix after fracture test, showing the good fibre–matrix adhesion [88].

Tables 3 and 4 gather the most relevant results of single fibre tensile tests realised with rCFs and rGFs. It can be seen that solvolysis has a smaller effect than pyrolysis on the mechanical properties of both CFs and GFs. However in strong solvolysis conditions (oxidant, acidic or alkaline), CFs can be seriously damaged as well. GFs are much affected by the treatment temperature in particular, but it appears possible to find some solvolysis conditions in which their degradation can be minimised even if they are still significantly damaged compared to virgin fibres [71]. The global tendency for both glass and carbon fibres is that the lower the temperature the lower their degradation.

It has been observed that no deep understanding of the chemistry involved in the solvolysis of resins has been realised in the literature. And yet a successful application of solvolysis at HTP with water or other solvents as reaction media requires the right combination of the chemistry involved in the resin degradation with that of the solvent properties provided at HTP. Indeed Elghazzaoui [72] observed that the epoxy resin of Hexply 914 cured prepreg was more easily degraded in subcritical conditions than in supercritical conditions of water, with the latter conditions leaving the recovered fibres coated with resin residues and glued to one another. The degradation pathway of UPs in subcritical HTP hydrolysis at about 275 °C has been shown to be an $A_{ac}2$ mechanism (acid auto-catalysed acyl-bond breaking of ester functions) [110]. Under these conditions the ionic product reaches its maximum and the reaction mechanism is similar to that for model molecules or for simple esters at ambient temperature. The main secondary reactions were also identified: decarboxylation of produced dicarboxylic acids and dehydration of glycols, which were also acid-catalysed. Fully hydrocarbon compounds are generally resistant to HTP hydrolysis. Compounds particularly susceptible to HTP hydrolysis are those containing a saturated carbon atom attached to a heteroatom-containing functional group [116]. The hydrolysis mechanism of epoxy or PEEK resins at HTP have not been much investigated, however we think that C–O, C–N, or O=S=O bonds, that are heteroatom-containing bonds in epoxy resins, are hydrolysable. The amino-alcohol group (red circle in Fig. 5) as well as the $-SO_2$ group (blue circle in Fig. 5) are the main hydrophilic sites in these epoxy-based materials [117]. The chemical structure of PEEK resin, shown in Fig. 6, presents C–O bonds that are hydrolysable

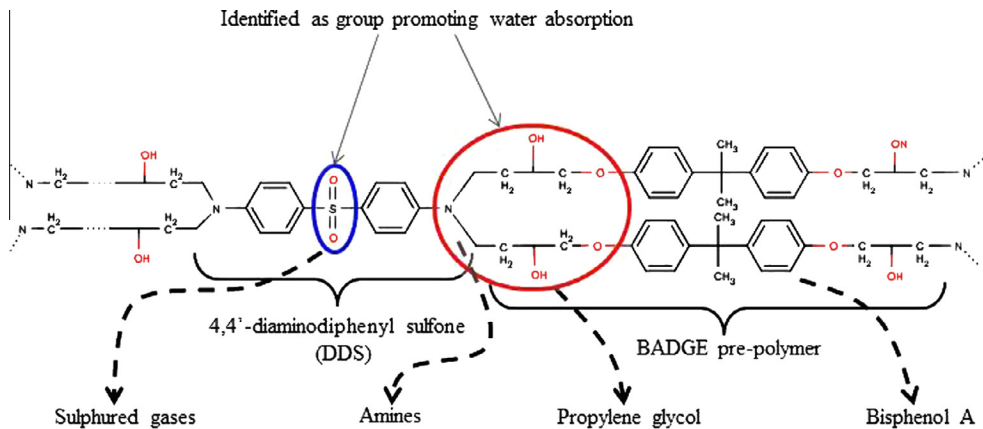


Fig. 5. Example of epoxy resin network obtained with BPA and DDS as curing agent – HTP hydrolysis might theoretically produce sulphured gases, amines, propylene glycol and BPA.

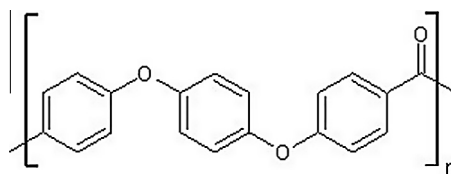


Fig. 6. Chemical structure of the PEEK resin.

but the steric hindrance is such that they are actually harder to break, explaining why a higher solvolysis temperature is required. However when the solvent is not water, no mechanisms have been proposed. Solvolysis at HTP is consistent with accelerated ageing, in particular when the solvent is water. Work on hydrothermal ageing [118–120] has shown that water diffusion in a resin and in a composite material and the subsequent resin degradation are influenced by interactions of water molecules with the resin's hydrophilic sites and also with the fibre–matrix interface (sizing). Water can build relatively strong hydrogen bonds with polar groups in the resin network, in particular between groups in close proximity to form a complex with a water molecule. In all inventoried studies about hydrolysis, the water was always de-ionised or distilled but at industrial scale the amount of solvent would be greater so mains water might preferably be used. Laboratory-scale experiments must therefore use mains water to confirm the industrial scale operation.

Post-treatment of recovered organic fractions has also received very little attention in the inventoried studies, in terms of recovery of valuable products or just recycling as they may contain toxic substances. If the objective is to recover only fibres, and to dispose of the organic fractions in energy recovery, then pyrolysis would appear as the more economical technique.

We have also observed that no mixtures of resin type or composites have been studied in solvolysis, for example epoxide with polyester and/or thermoplastics, depending on the reinforcement. It might be interesting to investigate if a synergistic effect could arise from the degradation of resin mixtures, as well as to consider resin mixtures reinforced with different types of fibres. A mixture of glass and carbon fibres for example might be interesting in certain applications, where a replacement of vGFs by rCFs was tried (presented in Section 3.2 hereafter).

Finally, all the existing techniques to recycle composites have advantages and drawbacks. All the inventoried work showed that it was possible to obtain grades of differing quality of fibres depending on the conditions. However a trend emerges: mechanical grinding is more suited for GFRCs and pyrolysis or solvolysis for CFRCs, when the damage to GFs by thermo-chemical processes and the potential commercial value of rCFs are considered. According to ATI [58], pyrolysis, HTP and LTP solvolysis have decisive drawbacks; however combined together they can produce rCFs of optimum quality. For this reason they have developed a three step process, which included a thermal pre-treatment followed by two solvolysis steps, first at LTP and then at HTP if resin residues remain on the fibres.

3. Review of reuse applications and discussion

3.1. Products from the resin

Products from resins are mainly recovered from thermolysis and solvolysis. Thermolysis produces gases and oil from resin degradation. Pyrolysis of glass fibre reinforced SMCs, that initially contain only about 25 wt.% resin, produced about 75 wt.% solid residue, 14 wt.% oil and the remainder as gases [20]. The oil recovered from the degradation of polyester resin contains monomers that could be reused in a new resin [21], providing a cost-effective recovery method is used. However material recovery from the polymer was judged not economically viable [34]. This condensable product is a complex mixture of numerous different compounds, mainly aromatics due to the presence of styrene and phthalic acid in the resin structure, with a broad spread of boiling points and a high content of oxygen. It might be difficult therefore to separate them according to their boiling temperature. Carboxylic acids like phthalic or isophthalic acids, which are main monomers of polyesters, could be easily separated during condensation as they are solid crystalline products [21]. Furthermore, not all the products are valuable, and the amount of valuable products should be present in sufficient amount to justify their separation. The remaining solution of non-valuable products would still require disposal. The energy recovery therefore seems to be the most suitable solution up to now to reuse the liquid fraction. The liquid fraction has a gross calorific value (GCV) estimated around 32–37 MJ/kg, which is low compared to conventional hydrocarbon liquid fuels and is due to the high oxygen content [13,20,21,122]. Once the lighter components have been distilled, it could be reused as a fraction for blending with petrol [20,21]. Without post-treatment refining the composition of this oil

led to a closed-cup flashpoint below the limits specified by both UK and US health and safety legislation. Users would have therefore to face extra safety obligations in order to use it [21]. After distilling off the lighter volatile compounds, less than 70 wt.% of the oil with a flashpoint greater than 55 °C is recovered and about 40 wt.% of the oil was in the distillation range equivalent to petroleum [20,21]. The remaining 60 wt.% could be reused as commercial heating fuel; however the oil fraction represents only about 14 wt.% of the initial amount of composite waste. After pyrolysis and post-treatment less than 10 wt.% of the composite is recoverable as fuel. The gases were mainly CO₂ and CO for pyrolysis of polyesters and provided a low GCV as well, nevertheless high enough to heat the process [20,119]. Pyrolysis of composites made of epoxy resins produced gases rich in methane, with a high GCV (between 42 and 44 MJ/kg) [13]. The condensable products for such composites have been less studied as they generally contain carbon fibres that have higher value. The degradation of epoxy produced mainly aniline in the liquid condensable products and water in the gaseous products [13]. In a fluidised-bed process only gases are recovered from the resin, with a composition similar to the one observed for pyrolysis. It is assumed that the gases produced might be used for self-heating the process, but was not considered in the literature.

Solvolytic produces mainly a liquid fraction from the resin degradation, in which products are dissolved according to their solubility. Depending on the solvent used, it is possible to observe sedimentation of components that are not soluble or form due to saturation at ambient conditions. Depending on the conditions small amounts of gases were produced, showing the same composition as those found with pyrolysis [72]. The interest in the use of a solvent is that thermal reactions occur, which can reduce the production of gas, tar and coke [122]. The solvent is generally a reactant and dissolves the products of the reaction, preventing or delaying higher order reactions just by dilution [122] and thus potentially enabling the recovery of resin monomers or at least more valuable products than with pyrolysis. It has been shown that hydrolysis of polyesters, for example, was the reverse reaction of esterification and leads to the production of resin monomers (glycols and carboxylic acids) and of a styrene–fumaric copolymer (SFC) resulting from the styrene network created during polymerisation [67,109]. Glycols were separated from the liquid fraction and reused in a new UP and the SFC was modified to a low profile additive (LPA) by using 1-octanol and sulphuric acid (H₂SO₄) [66]. The LPA thus obtained was successfully incorporated in a new GFRC. A similar linear polystyrene derivative was also recovered from UP by hydrolysis in presence of an aminoalcohol [123]. A post-reaction was performed with maleic anhydride (MA) and the resulting functionalised polystyrene was then crosslinked with styrene to give a solid polystyrene-based polymer. The latter was submitted to the same hydrolysis treatment to give back the linear polystyrene derivative. The authors do not suggest any applications; however we may assume that the obtained polymer could replace existing polystyrene-based materials. Another work [124] enabled the manufacture of an unsaturated polyester resin by the recovery of dimethyl phthalate (DMP) from GFRC by solvolysis in supercritical methanol in presence of N,N-dimethylaminopyridine (DMAP). Black oil was separated from methanol and contained about 31 wt.% DMP, which was then recovered and purified by washing with water. A new polyester resin was manufactured with the purified DMP, ethylene glycol and maleic anhydride and crosslinked with styrene. The first results were not satisfactory because of the presence of DMAP in the oil, but after purification the authors successfully manufactured samples containing different proportions of DMP with hardness comparable to that of the resin without recovered DMP.

Less work has been done on reuse of products recovered from epoxy resin. Globally epoxy resins cured with amines lead to phenolic mixtures [51,59,72,74,78,125]. Only one work realised by ATI displayed interest in the reuse of this mixture [125]. The recycling process developed by ATI produced a mixture that the authors found to be similar to the complex mixtures of phenols recovered from biomass pyrolysis. The work showed that the mixture could potentially be used to produce phenolic resin. However this was performed with a mixture made of model compounds and not on a real one. A hand-made epoxy resin crosslinked with a dicarboxylic anhydride was solvolysed in supercritical methanol [73]. The ester bonds created were easily broken by methanol and the epoxy network was preserved. The recovered thermoplastic epoxy (black viscous oil) was then re-crosslinked with acid anhydride.

Reuse of products from polyester degradation has been more considered in the knowledge that rGFs alone could not be expected to make a separation process viable economically. For CFRC, carbon fibres are so valuable that products recovery from resins has almost been neglected. The only considered solution was energy recovery. Due to the broad range of resin formulations, organic fractions recovered by either thermolysis or solvolysis may lead to very complex mixtures of products. However it was shown that ortho- and isophthalic polyester resins, which represent more than 95% of the total volume of GFRC, could be treated together as they have similar behaviour towards hydrolysis; whereas a specialist UP modified by dicyclopentadiene showed a different behaviour and was reported to require a specific treatment [110]. It also appears to be necessary to consider epoxy resins according to their curing agent. A classification is necessary therefore according to resin structure and behaviour towards separation processes, which requires an understanding of the reaction mechanisms. Thermoplastic resins like PEEK would similarly necessitate a classification. Whatever the chosen solution, energy or product recovery, reuse is not straightforward and requires separation and purification steps. The few studies realised up to now have shown significant potential in the reuse of products from resins. Further investigations and developments are necessary to propose suitable recovery solutions. In light of this, new resins that are recyclable have started to be developed, in particular by Adesso Advanced Materials in Japan [126] and Connora Technologies in USA [127]. Adesso proposes either recyclable curing agent for epoxy resins or recyclable epoxy systems. Connora has developed a recyclable curing agent for existing epoxy systems and a low-energy based recycling solution that produces after solvolysis treatment an epoxy thermoplastic by breaking a specific bond in the crosslinking agent. They are also working on the development of a crosslinking agent for polyesters.

3.2. Fibres: glass, carbon and aramid

Different types of fibre waste exist according to the step in the manufacturing process. Dry fibre waste inherent in the production of reinforcements is generally already treated by manufacturers such as Hexcel and Toray. Fibre waste, either dry or wet, is produced during the first steps of the production of composite parts. It can also arise from prepreg rolls that did not pass the quality control, that stayed out of the recommended storage conditions for too long and those outside the guaranteed shelf life. After a part is manufactured, finishing steps produce processed material waste that cannot be directly reused, in particular waste from thermosets; some parts can be scrapped after quality control.

A significant amount of waste is produced during these steps. According to Alex Edge from ELGCF, the majority of the carbon fibre waste they treat actually arises from these steps and not from end-of-life carbon fibre composite parts. This is due to the long service life of these materials, but also certainly because the waste stream has to be implemented between the dismantling sites and the recyclers.

3.2.1. Fibrous fractions recovered from mechanical recycling

Ground carbon and Kevlar[®] fibres from cured thermoset moulded parts have been incorporated into sporting goods prototypes like snow and water skis [2]. Carbon fibres gave better improvement in the mechanical properties of the resulting material than Kevlar[®] fibres. Experiments showed that when the amount and/or length of fibres were too high, the dough was too viscous, difficult to mix and the resin did not uniformly impregnate the ground fibres. The best result was achieved with fibres of 0.5 mm length at a loading of 1% in an epoxy resin, giving a strength increase of 16%. The incorporation in polyurethane foam (used in skis, wall and pipe insulation, and the packaging industry) at a maximum amount of 0.5% gave approximately 15% improvement against failure under load. It was indicated that the two-step process cost was estimated to less than \$2 per pound of recycled material in 1995.

Carbon fibres, as well as glass and aramid fibres, recovered from a grinding and sifting process were also characterised by Kouparitsas et al. [15] and the fibre-rich fractions were reincorporated in thermoplastic resins. The incorporation of ground GF in polypropylene (PP) at 40 wt.% and of ground aramid fibres in ionomer resin at 15 wt.% gave tensile strength comparable to the same resins

reinforced with virgin fibres. In contrast ground CFs incorporated at 20 wt.% in ionomer showed a tensile strength reduced by about 35% compared to the same resin reinforced with vCFs.

More recently ground thermoset CFRCs were reincorporated in new SMCs by Palmer et al. [18]. A classification and sieving method enabled separation of the products into four grades. One grade contained small, fine bundles of fibres approximately 5–10 mm in length. They were similar in length and stiffness to glass fibre bundles normally used in automotive SMC. This fraction corresponded to 24 wt.% of the recycle and showed a fibre content of 72 wt.%. About 20 wt.% of the vGFs were replaced by this grade of carbon fibre recycle. The obtained SMC composite showed mechanical properties comparable to that of the class-A standard automotive grades and good surface finish. However, at the end only a quarter of the original material could be reused and at a replacement yield of 20 wt.% in a new material.

Takahashi et al. [16] crushed thermoset CFRCs into square flakes of about 1 cm² that were then incorporated into thermoplastic resins to manufacture materials by injection moulding (acrylonitrile–butadiene–styrene ABS and polypropylene PP). Compared to the unreinforced resin, the material with 30 vol.% crushed CFRCs showed better mechanical properties except for the flexural strain at fracture, and even comparable to ABS reinforced with the same amount of vCFs. The same tendency was observed with PP reinforced with crushed CFRCs, and the authors also showed that repeating four times the injection moulding process did not affect significantly the mechanical properties. The obtained properties appeared to be comparable to current glass fibre reinforced thermoplastics (GFRTPs).

Similarly, Ogi et al. [17] also crushed thermoset CFRCs into small pieces with average size of 3.4 mm × 0.4 mm and further ground some into 1–10 μm particles. The mechanical characterisation showed that crushed CFRCs provide a better reinforcement than milled CFRCs thanks to longer fibres. The incorporation of crushed CFRCs showed an optimum level (50 wt.%) above which the mechanical properties deteriorated with increasing content. According to the authors, the crushing process would have removed epoxy resin from a part of the fibres which enabled them to be coated with the thermoplastic resin. However the nonlinear stress–strain curves obtained at different fibre contents were attributed to microscopic damage during the testing due to debonding and resin cracking. This would mean that the adhesion of the thermoplastic resin with the fibres was globally the weak point of the material. The fibres also seemed to be somewhat oriented in the direction of the injection flow and present as single fibres as well as fibre bundles in the material. Other work also exploited the unique properties of this type of recycle [13,91]: as a core made of ground glass fibre composites, for example.

Ground CF reinforced PEEK resin (C/PEEK) incorporated into virgin PEEK resin in different weight proportions and processed by injection moulding gave a material with mechanical properties comparable (even better) than the same virgin material [14]. The elastic modulus increased with the proportion of ground C/PEEK (+60% from 30 to 50 wt.%), while the tensile strength decreased but to a low extent (first increase of 2–15% from 30 to 40 wt.% and then decreased by 6–8% from 40 to 50 wt.%). The strain capacity was more affected by the ground C/PEEK content: it was almost not affected from 30 to 40 wt.% but from 40 to 50 wt.% a decrease of 18–36% was observed. Press moulding of C/PEEK without adding virgin PEEK resin led to improved flexural properties compared to the same virgin material (up to 18% at 5 bar); however the higher the moulding pressure, the poorer the flexural properties. This might be explained by less resin between the fibres due to a spinning-like phenomenon. Finally, the same C/PEEK was directly reformed without grinding post-treatment. This led to a material with almost unchanged mechanical properties [14]. This was not undertaken with end-of-life materials but it may be appropriate for production waste. Pull-out testing would also be necessary in order to analyse the mechanical bonding between the resin residue on the fibre and the new resin.

Fibrous fractions recovered from mechanical methods appear to be more suitable for reuse in bulk or sheet moulding compounds, however they actually seem to better suit incorporation into thermoplastic resins, whether initially reinforced or not. This might be due to a better adhesion to TP resins as their processing does not rely on a chemical reaction to obtain the final material and thus the adhesion is more mechanical than chemical. Considering the effect of this incorporation on the resulting material properties, such recycled fibres cannot be reused in structural applications.

3.2.2. Fibres recovered from thermolysis and solvolysis

3.2.2.1. Glass fibres. Glass fibres recovered by fluidised-bed pyrolysis at 450 °C [23] showed a tensile strength reduced by 50% while at 550 °C the reduction reached 80%. It was however possible to restore some fibre strength with a potassium salt at high temperature. Potential applications of such fibres are limited by their discontinuous and fluffy nature. Incorporation of glass fibres reclaimed from SMC into DMC for compression moulding did not affect tensile, flexural or impact properties at concentrations up to 50%, but beyond this percentage all properties significantly deteriorated [23]. According to the authors, light duty parts such as vehicle headlight housing and instrumentation panels offer significant potential for commercial applications.

The University of Strathclyde announced a patent application covering cost effective, industrially applicable, treatments to regenerate the strength of thermally recycled glass fibres [128]. This could increase the value of rGF and widen their potential applications, but it will also increase their cost, which may not be competitive compared to vGF.

Glass fibres recovered from HTP hydrolysis [113] were incorporated into BMC using two different methods to improve the adhesion to the new resin: the refunctionalisation of the rGF and the incorporation of coupling agents directly into BMC semi-products. The results showed that both methods could improve the adhesion between the rGF and the new resin (Fig. 7).

The maximum fibre reincorporation percentage was 20% in order not to induce a significant decrease in the mechanical properties (the report does not indicate whether it was mass or volume %). Real parts were successfully manufactured without changing any process parameters; however the surface finish was of lower quality.

GFs recovered from thermolysis at 550 °C were reused to successfully produce a glass–ceramic material that could find applications in architecture [121]. The factor of interest is that separation is not necessary for GFRC containing fillers, such as calcium carbonate, as they can be processed with the recovered GF.

In light of the above, it appears that GF recovered by thermolysis or solvolysis does not allow a reuse of higher value than that for GF recovered by mechanical methods. Considering the price of those methods, which is evidently higher than that of mechanical grinding, we might conclude that such fibre–matrix separation techniques are not suitable for GFRC. It might be worth using these techniques only on composite materials reinforced with long high value GF, but this needs to be demonstrated.

3.2.2.2. Carbon fibres. Carbon fibres recovered by thermo-chemical techniques have been more considered for reusing in new composite materials (see Table 6). Random and aligned discontinuous fibres have been investigated [72,76,86–97] but very few real prototypes have been manufactured (Table 7). Furthermore, two types of rCF can be identified: random short fibres from either woven or non-woven fibre composites and pieces of woven fabrics. The size of recovered pieces of woven fabrics depends on the shredding pre-treatment; however the fabrics can actually retain their woven shape after the recycling treatment. This can be very interesting in terms of fibre alignment and so in terms of reinforcement retention. This type of reinforcement has received little attention [76,88,94], whereas it represents more than 60% of CF waste just in Europe [76]. Most often rCFs are not woven; in a wide range of lengths the fibres are intermingled like a tuft of hair. Different trials have been performed using rCF either as is or reshaped by different techniques (wet papermaking method, dispersion in liquid medium or by compressed air...). The other important issue is the absence of sizing on the recycled fibre surface and the presence of residues that can coat the fibres.

Due to their short length, rCFs were first incorporated into random discontinuous fibre materials like SMCs and BMCs. They generally need to be chopped again after reclamation in order to conform to virgin fibre length commonly used for this type of material [88,95]. Some trials used rCFs as they were [61,65], but the resulting materials were not homogenous in fibre distribution, thus in thickness, resulting in a quite low fibre content and high void content. Despite a fibre content lower than that of a BMC with vCF, the flexural properties were equivalent to the virgin BMC [72]: ultimate tensile strength decreased by 5% and flexural modulus decreased by 15%. The flexural properties obtained for an SMC were between 87% and 98% of those obtained for the SMC with vCFs, fibre contents being comparable [72].

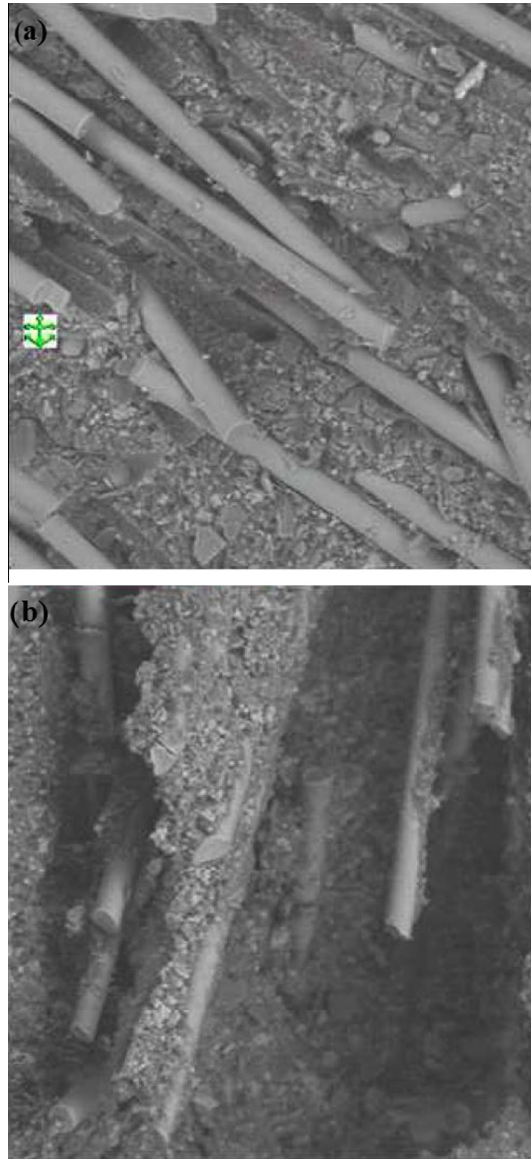


Fig. 7. SEM images showing adhesion between recycled fibres and matrix in a new BMC product: (a) bad adhesion without coupling agent and (b) improved adhesion with coupling agent [113].

Other methods [88,92,96] used two manufacturing processes, compounding and injection moulding, to manufacture BMC materials. If the rCFs were too long, they gave pellets that were difficult to process for BMC [88]. The materials were made with TP resins (polycarbonate [88] – PC, polypropylene [92] – PP, and polyphenylene sulphide [96] – PPS) and rCFs from pyrolysis (ELGCF process [88], FBP [92] and MIT-LLC process [96]) and from solvolysis (ATI process [88]). The material containing ELGCF rCFs showed properties very comparable to the material with vCFs (Fig. 8). ATI rCF materials showed an average decrease of about 36% in stiffness and strength properties; however the material was harder to process because of the longer fibres, but probably also because the fibres were coated

Table 6

Composite materials manufactured with rCF for mechanical testing.

Company/organisation	Country	Products
North Carolina State University [88,90]	USA	Injection moulded compounds with rCF from ELGCF and ATI and polycarbonate, rCF reinforced plastics by RTM: fibre mats then a preform. The wetlay process, commonly used for making paper
Université du Quebec Montreal, Bell Helicopter Textron Canada and NRCC [96]	Canada	rCF mats, then used to manufacture infusion/compression moulded composites Pellets of PPS reinforced with rCF moulded by injection
FibreCycle project [93]	UK	rCFs (length >500 mm) blended with thermoplastic staple fibres into yarns
University of Nottingham [92]	UK	CF veils to manufacture electromagnetic interference shielding by RTM PP/rCF by compounding and injection moulding, with and without coupling agents
University of Warwick, Umeco Composites Structural Materials, Lola group and ELG Carbon Fibre [95]	UK	Woven CFs reused in high performance energy absorption structures University of Warwick: reuse of woven rCF to manufacture the rear of WorldFirst F3 car
National Technical University of Athens [15]	Greece	Ground CFR epoxy in ionomer Ground GFR polyester in polypropylene Ground aramid fibre reinforced epoxy in ionomer
Ehime University and Toray Industries Inc. [17]	Japan	ABS reinforced with crushed CFRC by injection moulding (fibre length about 200 µm)
University of Tokyo and Toray Industries Inc. [16]	Japan	ABS and PP reinforced with crushed CFRC by injection (CFRC square flakes of about 1 cm)
University of Nantes, ICAM and Compositec [72]	France	Realigned rCF to manufacture UD coupons BMC and SMC
Imperial College of London [95]	UK	rCF recovered from uncured CF/epoxy fabrics by ELGCF pyrolysis and reused in a laminate made by resin film infusion (RFI)
Luleå University of Technology and Swerea SICOMP [91]	Sweden	rCF reused in preforms (mat, by papermaking method) then laid up with films of PP scrap and hot press formed
Saxon Textile Research Institute [97]	Germany	Non-woven rCF fabrics (stich-bonded or needle-punched) suitable for medium-strength requiring applications
University of Seattle, Automobili Lamborghini and Japan Ministry of Defense [76]	USA, Italie and Japan	Entangled rCF randomly distributed recovered from woven fabrics, however with a majority of fibres still in tow shape. The initial length of the fibres is retained. Remanufacturing by direct incorporation and VaRTM
University of Ulster [98]	UK	Polymer composite materials made up of waste CFs (collected from a 3D weaving process) and manufactured by injection and compression moulding

Table 7

Real part demonstrator and commercially available semi-products made of rCF.

Company/organisation	Country	Products	
Materials Innovation Technologies RCF (MIT-RCF) [86]	USA	Preforms and finished parts by the 3-DEP process (for example front lower wheelhouse support for the Corvette)	Real part demonstrator
Janicki Industries and Boeing [95]	USA	A tool for composite lay-up	
Technical Fibre Products (TFP) [129]	UK	Carbon veils (Optiveil™ eco) and mats (Optimat® eco)	Commercially available
Sigmatex Ltd. [130]	UK	Commingled thermoplastic fabric based on rCF and PET matrix	

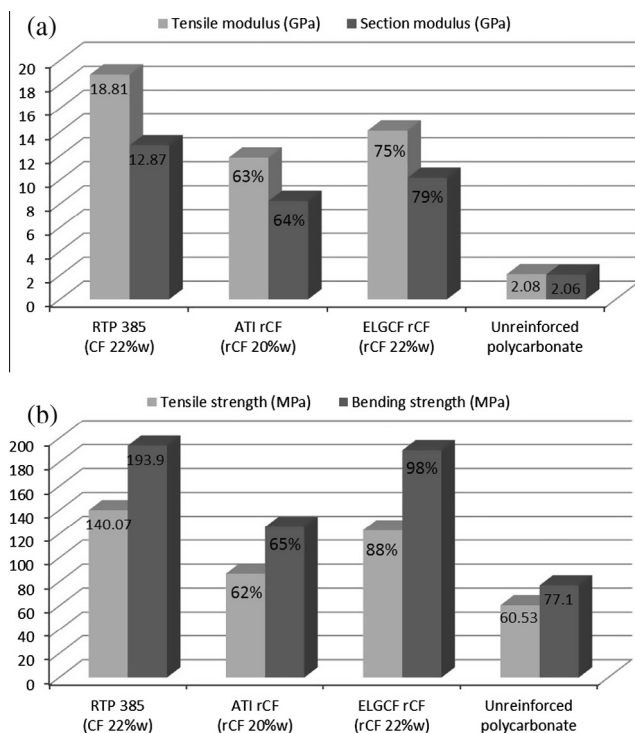


Fig. 8. Modulus and strengths measured from polycarbonate resins unreinforced and reinforced with either rCFs from ELGCF and ATI or vCFs [88].

with resin and catalyst residues, and insufficiently dried. The measurement of resistivity for both polycarbonates reinforced with vCFs and with rCFs revealed that the fibres are more randomly distributed in the material containing rCFs. Compared to the unreinforced resin, the incorporation of CFs, either virgin or recovered by pyrolysis, enabled an almost doubling in the tensile and bending strengths. This confirmed the reinforcement potentiality of rCFs.

The properties of the PPS materials were compared to those of equivalent composites containing vCFs. The authors [96] observed that the rCFs showed a broader distribution in single fibre tensile modulus and strength due to the various types of materials (each made from different CF grade) that can be found in feedstock. Consequently, the average mechanical properties of PPS composites containing rCFs were not as good as those obtained with vCFs. However the rCFs/PPS materials were completely comparable to existing PPS resins reinforced with short vCFs, and in some cases slightly better (Fig. 9). rCFs were essentially aligned along the injection direction and homogeneously distributed in the matrix, similar to the commercial materials. Despite the absence of sizing on the rCFs, good fibre-matrix adhesion was observed giving a failure more by fibre rupture than by fibre pull-out. The replacement would be rated a complete success if the rCFs were cheaper than the short vCFs used, which are low cost fibres. In terms of Life Cycle Assessment (LCA), it may be assumed that the environmental impact is in favour of the rCF for this type of material. These aspects are further discussed in the next section.

Mats and veils have been manufactured by wet papermaking methods [89,91,95]. The method induced a preferred orientation in the mats with 60% of fibres having the same orientation in plane and a very small proportion in out-of-plane direction [95]. The mats were then used to manufacture composite laminates by compression moulding with films of either epoxy [95] or PP [91]. Mechanical testing showed that the epoxy materials obtained had specific mechanical behaviour especially in

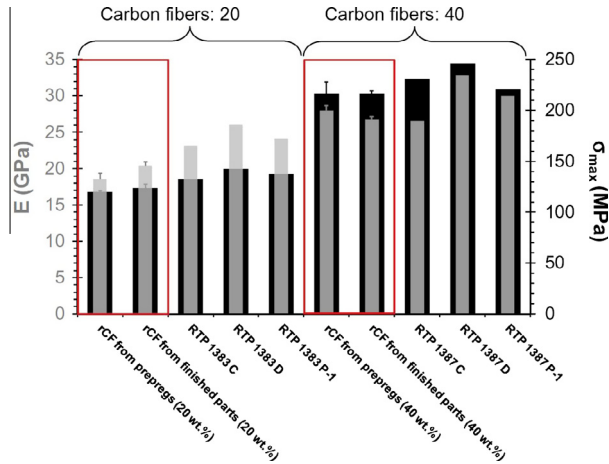


Fig. 9. Tensile properties of PPS composites reinforced with rCFs and commercial PPS composites reinforced with short vCF [96]. Young modulus in grey and tensile strength in black.

terms of failure mode [95]. The fibre–matrix adhesion was good but the tensile strengths were low. The extremely high moulding pressures required to manufacture rCFRCs with high fibre content led to severe breakage during compression, which degraded the fibre length considerably and left more than 60% of the fibres with a length lower than critical. Furthermore, the presence in the mats of fibre bundles (fibres that remained held together by resin residues or pyrolytic char) in the mats was shown to improve the local fracture toughness depending on their geometry, location and orientation. However, it seems that this was actually detrimental to the strength of rCFRCs. More investigations are necessary in order to improve the understanding of the mechanical response of rCFRCs. It was reported that TP materials gave a lower void content, probably due to better compression enabling improved resin diffusion [91]. Furthermore, the epoxy resin required a chemical reaction to achieve its final structure, which may have slowed resin diffusion into the mats. In both cases, fibre pull-out was observed, indicating a poor fibre–matrix adhesion.

The veils obtained by Wong et al. [89] were moulded onto a GF-reinforced polyester in order to provide electromagnetic interference shielding. Given that retention of fibre strength is not critical as in other applications, rCFs would be suitable for this type of application thanks to their low electrical resistivity. The shielding effectiveness increased with increasing veil area density of rCF up to 60 gsm. The performance could be improved by removing the long fibre portion from the rCF length distribution in order to achieve a more homogenous distribution.

As mentioned previously woven fabrics can be found among recovered CFs. Composites reusing woven rCF fabrics recovered from uncured prepreg by ELGCF pyrolysis were manufactured by Resin Film Infusion (RFI) at Imperial College London [95]. The pyrolysis conditions led to two types of plies: rCFs-B plies that lost their nominal rigidity and were thus more drapable, more deformed but also with significantly impaired mechanical properties (retention of only about 20% in single fibre tensile strength and visible defects); and to rCFs-D plies that kept their shape and nominal rigidity due to resin residues in the less aggressive conditions and showed a retention of 95% in single fibre tensile strength. When the shape and the rigidity of the plies were retained, the fibre re-impregnation by RFI was harder, and significant inter-tow and inter-fibre voids were observed in the composite, whereas the re-impregnation was better with no voids when the plies were more flexible. The autoclave manufacturing process improved the quality of the resulting material with fewer voids, but the compaction was still worse than the original material. The composite containing rCFs-B gave fibre content and elastic properties (tensile, compression and shear moduli) very similar to the original material, however resulting tensile strengths were significantly reduced (by 71–74%). On the other hand, the composite containing rCFs-D gave lower elastic moduli (average decrease of 15–20%), and

in particular, lower compression strength compared to the original material but also to the composite with rCFs-B (the rCF-D material strengths were decreased by 32–49% compared to those of the original material). The autoclave process improved the resulting properties but not sufficiently. The impaired mechanical properties would thus mainly be due to the lack of fibre wetting and resin diffusion into the rCFs-D plies during the remanufacturing process (the virgin material is a prepreg), but also to the residual resin on the fibres (about 7.6% of the original resin content) which prevented good adhesion to the new matrix. The new resin could not effectively penetrate the weave, all the more so, since the reclaimed fabric maintained its rigidity. Woven rCFs, which were also recovered from uncured prepreg by ELGCF pyrolysis, were reused by Meredith et al. [94]. The fabrics were re-impregnated and then all laid at 0°, vacuum bagged and cured in autoclave under pressure. The modulus of the obtained composite was slightly reduced compared to the same material with vCFs. Tensile and flexural strengths were significantly reduced, however the material retained 94% of its specific energy absorption capability, indicating that this type of material could be used in applications that require high performance energy absorption.

Carberry from Boeing [38] stated that the key to unlocking the manufacturing future for recycled fibre beyond injection moulded applications is aligning the fibres. Different methods have been tried, in particular by MIT-LLC [36,86,87] and by the University of Nottingham [36,95]. The 3-DEP process developed by MIT-LCC [87] enables to make three-dimensional preforms and is able to control the fibre placement and orientation. A demonstrator of the front lower wheelhouse support for the Corvette, currently manufactured with GFs was produced [36,86]. The materials obtained would be suitable for structural but not safety-critical applications. The University of Nottingham have also developed two methods for realigning the fibres: one is based on a modified papermaking technique [95] and the other is a centrifugal alignment rig [38]. They enabled the achievement of up to 80% and up to 90% of realignment respectively, however they were used to produce quite low density veils and mats. Very recently, at the University of Bordeaux, a process to unweave and realign the recovered tows was developed [131]. It enabled them to produce 50 mm wide tapes with aligned discontinuous fibres and an areal density of about 600 g/m². A prototype was built and is reported to be able to treat pieces of 200 mm × 500 mm with a yield of 3 kg/h. However the process requires improvement in terms of realignment quality and homogeneity of the area density. It can be concluded that there is no satisfactory realignment process and this issue is still in progress.

In order to really benefit from the reinforcement capability of rCFs, the other possibility would be to reshape the rCFs in continuous yarns. This has been done in the Fibre Cycle project [93], in which yarns were produced using rCFs recovered by ELGCF pyrolysis comingled with polypropylene (PP) fibres made by existing textile techniques. Only fibres of length higher than 500 mm were considered and then cut to 50–55 mm to be processed. They were subsequently blended with long PP fibres at different rCF/PP ratios. The crimped PP fibres acted as carriers for the un-crimped rCFs, providing the required fibre-to-fibre cohesion. Wrap-spun yarns of about 1 k tex linear density were obtained and wound onto a steel frame unidirectionally. The layers were then compressed and consolidated using hot compression moulding. The characterisation of the obtained yarns indicated a rCF content of 25 wt.% and 40 wt.% for ratios of 30:70 and 50:50, respectively. Fibre breaking occurred during the carding process, which led to lower fibre content than that of the initial blend. Most of the fibres showed a length decreased to 15 mm and down to 5 mm. Furthermore, the higher the initial rCF/PP ratio, the greater the breaking of fibres during carding, however higher ratios gave higher tensile and flexural strengths and moduli (about 1.5 times higher). After compression moulding, the rCFs were spread out uniformly in the PP matrix without evidence of voids. It also appeared that the majority of the rCFs (about 90%) were parallel and aligned with the yarn axis as shown in Fig. 10a. Nevertheless, SEM analysis of the fracture surface indicated that failure occurred largely by fibre pull-out indicating a weak fibre–matrix adhesion (Fig. 10b). According to us, this might be explained by the presence of resin residues or char on the fibre surface, considering the results of the fibre characterisation prior to processing. The interface between this coating and the rCFs might be weak and furthermore the smooth aspect of the fibre surface might have been detrimental to mechanical adhesion to the PP matrix.

In the AERDECO project [72], the best method to separate and realign the rCFs was found to be dispersing the fibres by blowing with compressed air; carding was not efficient and broke the fibres. First

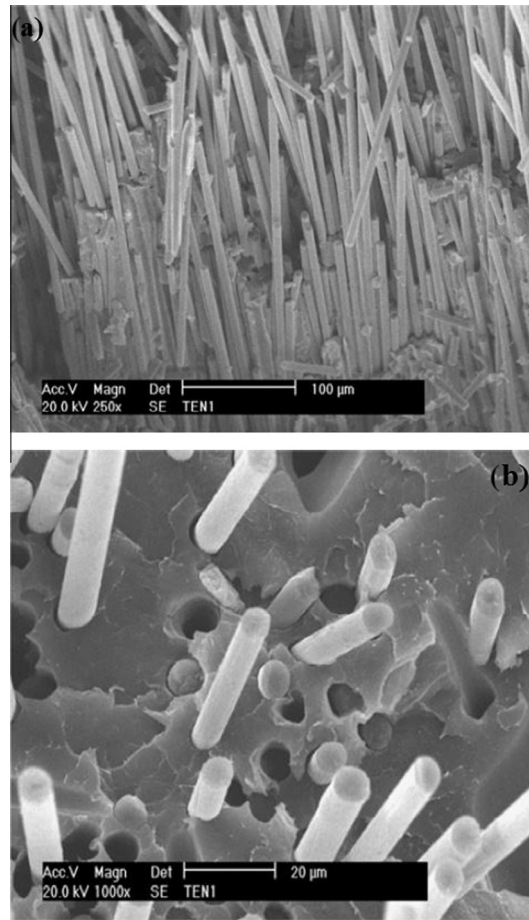


Fig. 10. SEM images of (a) rCF alignment in composite and (b) rCF pulled out from PP matrix [93].

trials of rCF reincorporation without post-treatment separation or surface treatment led to the absence of fibre–matrix adhesion, leading to the conclusion that residues were present on fibre surface. Realignment of long rCF bundles, found in batches of rCFs, was realised by hand to make unidirectional plies. They were then laid and vacuum moulded to make coupons. Due to the small size of the coupons, only bending tests could be performed and compared to real unidirectional laminates made with vCFs. The obtained flexural properties were almost the same as for the real unidirectional composite. This shows that rCFs are equivalent to vCFs in terms of reinforcement capability. At larger scale than that of the coupons, rCFs could enable the production of composite materials with aligned discontinuous rCF comparable to materials containing vCFs, but ensuring alignment, distribution and area density equivalent to a virgin material. This also shows that the advantages of rCF could be exploited if it is possible to make reinforcements with rCFs comparable to existing aligned discontinuous reinforcements (referred as stretch-broken fibres [132], and made by either stretching continuous fibres causing random breaking, or as continuous discontinuous fibres as shown in Fig. 11 [133]).

In all cases cited previously the surface quality appeared to be the weakness of the rCFs. Whatever the thermo-chemical method used to recover fibres, measurements of fibre diameter and observation and characterisation of fibre surface indicated that the rCFs were often coated by a layer of either char (more often seen after pyrolysis) or resin residues (more often after solvolysis). This resin residue

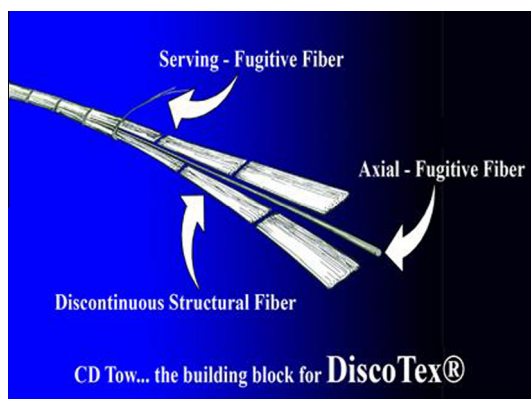


Fig. 11. Schematic of CD tow [133].

might consist of material from the fibre/matrix interface, comprising the sizing initially present on the fibre surface. As seen previously, this coating caused failures by fibre pull-out due to a weak fibre-to-matrix adhesion. For sized vCF, the surface oxygen content and the nature of the functional groups present on the fibre surface could influence the fibre–matrix adhesion. Different post-treatments have been tested in order to improve the fibre surface quality [92,108,113,133–136] but this has not been performed systematically. The objective was to modify the chemical structure of fibre surface in order to promote adhesion to a new resin either by oxidative thermal treatment, nitric acid treatment or plasma treatment. Greco et al. [108] studied different post-treatments, thermal and chemical. Thermal post-treatment was not satisfactory as it produced either more damaged fibres or fibres without evident surface activation, but fibre/matrix interfacial shear strength (IFSS) was improved by 40%. On the other hand, chemical treatment in nitric acid led to limited damage of the fibres while improving the surface chemistry (IFSS improved by about 70%). The use of DGEBA epoxy resin as sizing on rCFs treated by nitric acid [134] showed a significant improvement of the materials in which they were incorporated compared to the same material containing untreated rCFs. However, the harsh conditions of such post-treatments lead to fibre damage, sometimes even more than the reclamation process itself [108].

Work by Pimenta and Pinho [107] indicated that when the rCFs showed high surface purity the re-impregnation and adhesion to a new epoxy resin were better. This would indicate that re-sizing of the reclaimed fibres is not necessary before reuse.

A specific re-sizing of rCFs recovered by fluidised-bed pyrolysis [92] presented a very significant improvement of the mechanical properties of polypropylene composites and of the adhesion to polypropylene. Pimenta et al. [137] however, showed that the fibre–epoxy interfacial shear strengths were comparable for vCFs and rCFs recovered by pyrolysis at ELGCF, and in some cases it was even slightly higher for rCFs.

The presence of either striations or roughness seems to play a role in fibre–matrix adhesion and optimisation of mechanical properties [114,137]. Carbon fibres are furthermore known to be microporous and it was suggested that absorption of epoxy resin into these pores is a key factor determining interfacial bond strength in CF/epoxy composites [138]. However these pores may become clogged by organic residues in rCFs (presumably from the fibre/matrix interface). It was found that the presence of functional groups is not necessarily the most important factor, depending on the type of fibres [138]: for HM fibres removing the surface oxygen did not change their IFSS, whereas for HT fibres the IFSS follows the concentration of surface oxygen. In the case of epoxy resins reinforced by HM fibres, IFSS would depend more on the physical surface area which determines the strength of mechanical bonding [138]. A comparison of fibre diameters measured on vCFs and on rCFs recovered by subcritical and supercritical hydrolysis at 340 °C and 380 °C respectively [72] showed that the

recycled fibres were coated with an organic residue even after washing several times with solvents such as acetone or dichloromethane. Atomic Force Microscopy (AFM) confirmed that the surfaces of rCFs were not comparable to those of vCFs. The latter had smooth surfaces whereas the rCF's surfaces were rougher, even more so after supercritical hydrolysis. The remaining coating was then removed according to French standard NF EN ISO 10548, used to determine the sizing content of carbon fibres. The tensile modulus was halved compared to the value obtained before Soxhlet extraction. Similar behaviour is seen for glass fibres, where the modification of the structure of the fibres may be used to support this result.

Commercially, rCFs are available for reuse. Some applications have been proposed, but no products made of rCFs are available on the market. The studies carried out so far show that a direct reuse is difficult due to the fluffy character of the fibres after reclamation. In order to facilitate their reuse it might be necessary to develop semi-products similar to those made of virgin fibres to manufacture SMC, BMC, laminates, preforms, etc. The fibre re-alignment techniques require more development, as well as the techniques to make discontinuous fibre yarns. The characterisation of the fibre surface and of the fibre–matrix adhesion also requires further investigation. All those efforts are necessary for widespread industrial and customer acceptance [38]. If manufacturing costs were more affordable, the use of rCFs could attract interest in applications requiring less strength than for highly structural parts like those found in aircrafts or cars. Ideally rCFs should be incorporated in resins that are easier to recycle, for example TPs (polypropylene, polyamide, polyethylene terephthalate), or even in newly developed recyclable resins such as those mentioned previously in Section 3.1. Such materials could be used in the automotive industry, sports goods or niche markets like orthopaedic, laptops, mobile phones (Fig. 12) and even in aircraft interior components. Different grades of CF exist depending on the precursor and the carbonisation conditions. Most of the available CFs are based on polyacrylonitrile (referred as ex-PAN CFs). The carbonisation phase is used to enhance physical and chemical characteristics of the fibres and the conditions determine the carbon content of the fibres (Table 8). It would be relevant to classify the rCFs and related waste according to the fibre grade in order to optimise fibre reuse.

4. Discussion on economic and environmental aspects

Economic and environmental aspects have attracted more attention in the case of CFRC than for GFRC, because CFRCs have a larger impact on advanced engineering and contain more expensive fibres than GFRCs. However, if we consider the volumes consumed, CF composites represent only 1.5 wt.% of the world production, the vast majority being GFRC [102]. Nevertheless, the global demand for CFs is expected to double by 2020 [143].

All the studies on recycled GFRC lead to the conclusion that the best way to recycle these materials would be co-incineration in cement kilns; although significant controversy surrounds the environmental impact of using this strategy [144]. The price of vGF is so low that no process currently available can provide rGF with the same characteristics as virgin fibres at a competitive price. They would struggle to be used as fillers when considering the low cost of commonly used fillers; the economic balance is less clear than for CFRC. Two UK companies, Hambleside Danelow and Filon, are claiming that they can cost-effectively recycle GFRC through mechanical processes using waste from production [145]. When companies recycle their own waste in this way, the cost and the environmental impact are improved by the fact that there is no transport. The situation might be different when it concerns end-of-life waste. rGF are also too significantly damaged by thermochemical processes (thermolysis and solvolysis) to be used as reinforcement, even at low concentrations. Nevertheless, it might be relevant to distinguish short GF composites (SMC, DMC and other similar materials) from long GF composites. The latter can be found in boat building or even more in wind turbine blades [145] and are generally made with quite high value fibres. Investigations carried out in the EURECOMP project [113] in twelve west European countries revealed that boat building was the sector producing the largest volume of GFRC, generating production waste as well as existing end-of-life (EoL) waste (10 ktonnes/year). In modern cars the proportion of composite is relatively low [100].



Fig. 12. (a) CF orthosis [139], (b) laptop cover made with a blend of TP resins reinforced with CFs [140], (c) mobile phone shell [141].

Table 8

Fibre grades properties according to carbonisation temperature [142].

Carbonisation temperature (°C)	≤1000	1500–2000	≥2000
Grade of CF	Pre-Ox	IM	HM
Modulus of elasticity (GPa)	4	250–235	325–400
Carbon content (%)	62	90–95	98

The subcritical hydrolysis treatment developed to recycle GFRC in this project was also economically and environmentally evaluated by comparison to mechanical recycling and pyrolysis [113]. The study revealed that the developed solvolysis treatment would produce a rGF that is more expensive than vGF and showed an environmental efficiency lower than mechanical recycling but comparable to that of pyrolysis. The main drawback appeared to be the energy consumption of the process. Based upon scale-up of a pyrolysis pilot plant, Pickering et al. [23] evaluated the economic viability of their fluidised-bed process to recycle GFRCs. Their assumptions, including waste collection

and grinding post-treatment, led to a break-even point at a scrap capacity of about 9000 tonnes/year. This appeared a little unreasonable with assumptions of waste sourced within 80 km radius and of rGFs retaining 80% of their initial mechanical properties. Improvements are necessary to make these types of treatments applicable to GFRC. A first step could be the study of milder solvolysis conditions (lower temperature and pressure). In contrast, this is not possible for pyrolysis as a minimum temperature (≥ 450 °C) is required to volatilise the resins (as explained in Section 2.2). In order to make thermo-chemical recycling treatment more economical, it would also be necessary to find a way to recover valuable products from the resins. Resin prices follow the evolution of petroleum prices, so higher monomer prices might justify this approach. Due to the much higher price of vCF, solvolysis and pyrolysis processes appear to be more applicable to CFRC, at least in terms of the economics.

CFs cost more because they require more expensive raw materials, and moreover, more energy to be produced in comparison to GFs (Table 9). Furthermore, if we consider the manufacture of laminates using long CF prepregs, the high energy consumption is due to the energy necessary to produce the CFs (Table 9), then the prepregs and finally the composites by the autoclave process (Table 10). These kinds of materials are particularly and increasingly used in aeronautics, but their recycling will not generate rCFs that can be reused in the same applications, unless a recycling technique able to preserve CF characteristics (performance, length and architecture) is developed. Currently available fibres reclaimed from cured composites are generally unsized, limited in length (typically less than 100 mm) and available in random directional distribution, whereas virgin fibres are sized and available in every length between short and long, in unidirectional tapes or fabrics. rCFs cannot therefore be described as identical to vCFs. Reclaimed fibres could replace short virgin fibres in applications using discontinuous fibre composites. However rCFs are quite fluffy and thus difficult to manipulate. Although some studies have shown their reuse potential (as reported in Section 3.2.2.2 in this article), the proposed applications mainly concerned down-cycling or reuse

Table 9
Energy intensity for the production of fibres and resins [146].

Material	Energy intensity for production (MJ/kg)
<i>Resins</i>	
Polyester	63–78
Epoxy	76–80
PP	72–112
<i>Fibres</i>	
GF	13–32
CF	183–286
Flax fibre	6.5
<i>Metals</i>	
Aluminium	196–257
Stainless steel	110–210
Steel	30–60

Table 10
Energy intensity of manufacturing processes [146].

Process	Energy intensity for production (MJ/kg)
Autoclave moulding	21.9
RTM	12.8
SMC	3.5
Filament winding	2.7
Pultrusion	3.1
CF prepreg	40
Injection moulding	19

in unreinforced material. The incorporation of rCFs in such materials may lead to a higher cost and a lower environmental efficiency.

Boeing estimates that CFs can be recycled at approximately 70% of the cost to produce virgin fibres as given in Table 11. The prices for rCFs commercially available from ELGCF indeed are in this range (Table 12). It was suggested that vGFs or vCFs could be replaced by rCFs [34], for example in SMCs and BMCs. The resulting materials show a higher cost compared to vGF materials (Fig. 13a) even if the mechanical properties are better (Fig. 13b). Furthermore, considering the energy consumption of thermo-chemical processes it may also lead to a poor environmental balance.

Indeed a LCA study [149] revealed that when rCFs replace vGFs in proportions giving equivalent material stiffness, incineration with energy recovery (followed by landfilling) generally has a smaller impact. Whereas when rCFs replace vCFs with proportions giving equivalent material stiffness, recycling showed clear environmental benefits (Fig. 14). An analysis comparing CO₂ emissions related to hypothetical automotive components utilising vCF, rCF and vGF showed that the components using rCFs in replacement of vGFs can begin to be profitable, thanks to the weight gain, when travelling about 41,000 km. It is important to consider the component use life in evaluating impact on the environment and not only the manufacturing and recycling processes. Recycling by pyrolysis would therefore be the most environmentally favourable option for waste treatments compared to incineration and landfilling. Furthermore, it appears that when CF is recycled and reused to replace vCF, the benefit to the environment arises from avoiding vCF production impacts; similar conclusions were also reached by Hedlung-Åström [147]. The same study has to be done to evaluate the other available recycling techniques and compare them against each other, this is one objective of the EXHUME project [150]; this is also the objective of the French project SEARRCH [151]. It is necessary to define indicators to assess the economic and environmental impacts of each recycling technique (including incineration and grinding) for each type of material class and according to remanufacturing applications.

Finally unprocessed and out-of-date production waste (from fibre or semi-product manufacturers and from composite parts manufacturers) should be used without going through a recycling process in order to reduce the economic and environmental cost of their recycling. Unprocessed CF or GF have the advantage of still being sized, and are therefore easier to manipulate. They are also comparable to virgin fibres, also in terms of mechanical properties. Fibres of thermosetting prepreg waste could be separated by low temperature processes, in particular solvolysis, which could give improved recovery of valuable products from the uncured resin.

Table 11

Comparing virgin and recycled carbon fibre recovered by thermo-chemical processes [38] to ground CFRC [147].

	Cost to manufacture	
	Materials	Energy in kW h/kg (MJ/kg)
Virgin carbon fibre	US\$ 33–66/kg	55–165 (198–594)
Recycled carbon fibre (thermo-chemical recycling)	US\$ 13–19/kg	2.9–9.9 (10.3–35.7)
Ground CFRC	About US\$ 5/kg (3.36 €/kg)	0.074 (0.27)
Virgin glass fibre	About a tenth of the cost of virgin carbon fibre [145]	3.9–9.5 (13–32)

Table 12

Sales prices of rCFs recovered from pyrolysis at ELGCF (information kindly provided by ELGCF).

	Sales prices		
	Boxes	Pallets	Tonnes
Milled rCF or random chopped rCF	18 €/kg US\$ 25/kg	13 €/kg US\$ 18/kg	11 €/kg US\$ 15/kg

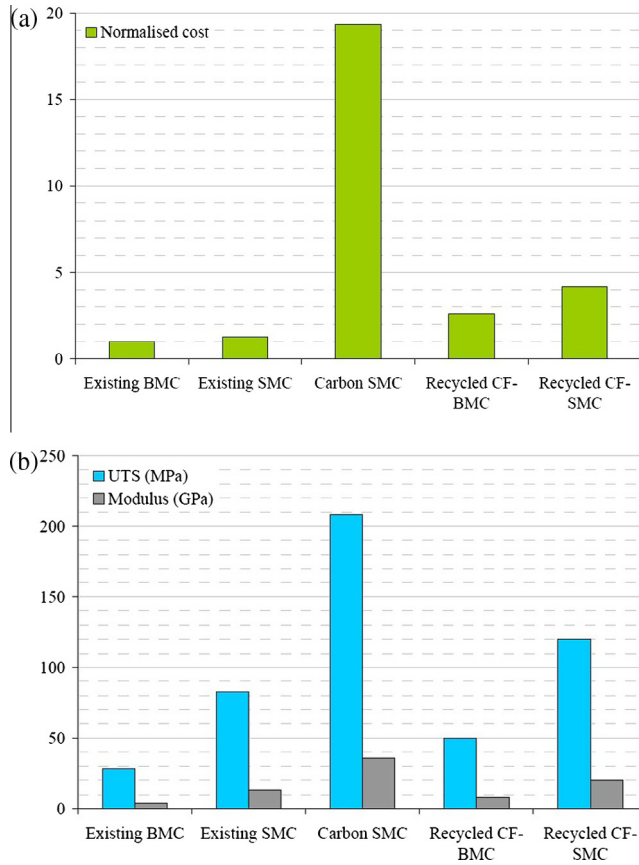


Fig. 13. Cost (a) and strength (b) comparison of existing SMCs and BMCs using GFs and SMCs and BMCs using rCFs [148].

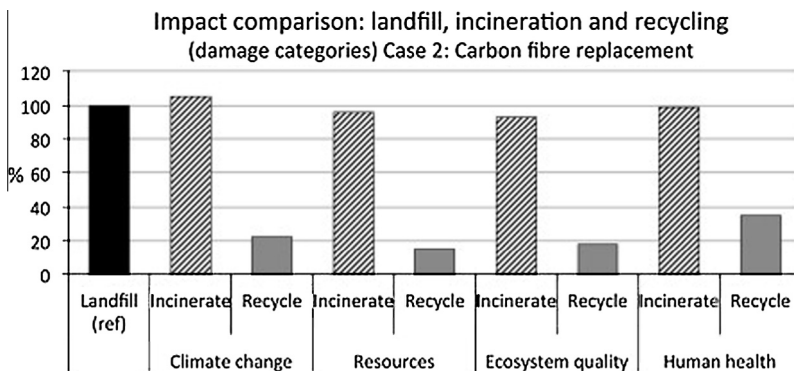


Fig. 14. Comparison of impacts from landfilling, incineration and recycling for the CF replacement case [149].

Table 13
Summary of the recycling solutions according to the type of processed composite waste.

	GFRC		CFRC	
	Thermosets (TSs)	Thermoplastics (TPs)	Thermosets	Thermoplastics
Recycling method	<ul style="list-style-type: none"> • Mechanical treatment • Cement kiln 	<ul style="list-style-type: none"> • Mechanical treatment • Direct re-processing 	<ul style="list-style-type: none"> • Mechanical treatment • Thermo-chemical treatment 	<ul style="list-style-type: none"> • Mechanical treatment • Thermo-chemical treatment • Direct reprocessing
Reuse methods	<ul style="list-style-type: none"> • Fibre-rich fractions reincorporated into TSs or TPs but limited proportions (≤ 20 wt.%) • Resin-rich fractions and powders as fillers or energy source 	<ul style="list-style-type: none"> • Reincorporation into the same resin after mechanical treatment • Direct reprocessing after or without mechanical treatment 	<ul style="list-style-type: none"> • Ground CFTSs in TS resins (injection or press moulding) but limited proportion, better results when incorporated in TP resins • Resin-rich fractions as fillers or energy source • CFs reclaimed from thermo-chemical processes as reinforcement in random discontinuous fibre composites (TS or TP resins); a re-sizing is preferable • Longer rCFs potentially reshaped into yarns (continuous discontinuous fibres) 	<ul style="list-style-type: none"> • Ground CFTPs incorporated into the same virgin resin and injection or press moulded (up to 50 wt.%) • Direct reprocessing • Resin-rich fractions as fillers or energy source

5. Conclusions

In the present article the most reported techniques to recycle composite materials were reviewed, these included both mechanical recycling and thermo-chemical processes (thermolysis and solvolysis). The inventoried studies led us to the conclusion that the recycling technique must be selected according to the material being recycled and also to the reuse applications. In light of this, a first classification arises as shown in [Table 13](#).

Mechanical recycling appears to be more suitable for GFRCS and possibly even for CF composites that are reinforced with either lower grade CFs and/or short CFs. This is currently done in-house by some manufacturers, in particular for GFRCS but this concerns only production waste. EoL waste is currently more often landfilled. The European composites industry has stated that the cement kiln route is the most suitable solution to dispose of GFRCS. We assume that this solution is also suitable to treat EoL GFRCS waste. Thermo-chemical processes are up to now not viable for GFRCS considering the low price of vGF and the degraded mechanical properties of GFs recovered in this way. These processes have been largely used to recover valuable products from the resin to reuse in new resins.

Most of the currently available CFRC waste comes from production. The durability of those materials and their recent rise will see this waste increase. However, EoL waste will be prevalent soon, largely from aerospace, and will require a proven recycling solution. Thermo-chemical processes are suitable for CFRC recycling due to the high value of the CFs. Pyrolysis is currently commercially exploited and two solvolysis processes are available for commercial exploitation. Both techniques are able to provide clean and high quality CFs; however they also consume more energy due to the high processing temperatures. These are hard to lower for pyrolysis whereas solvolysis can potentially be optimised. Both techniques have drawbacks and advantages and, according to the American company Adherent Technologies, are considered as complementary rather than competing. The best treatment for them is a combination of a wet chemical processing followed by pyrolysis to remove residual organic substance that may remain on the CF surface. A drawback common to both techniques however exists: the rCFs are fluffy in nature and show a specific surface quality (surface layer of residual fibre/matrix interface that is more or less rough). Due to this their reuse is hardly straightforward.

Depending on the type of rCF (HM or HT), it seems that the surface oxygen content is not always a parameter that influences adhesion in a new resin. In general a re-sizing step significantly increases the IFSS, regardless of the thermoset or thermoplastic matrix. It is particularly relevant when thermoset resins are chemically bonded to the fibres during the curing phase, whereas in thermoplastics fibre-to-matrix adhesion is also generated mechanically. In light of this the reuse of ground CFRTs in the same class of thermoplastic resin is very relevant. Furthermore, as CFs are short and randomly distributed after thermo-chemical reclamation, they cannot be used in highly structural applications, for example in parts from which they are recovered. rCF properties also strongly depend on the fibre length: the longer the fibres, the higher the probability of defects induced by the reclamation process, making the fibres more fragile. Composites are reinforced with CF of different types, and consequently the recovered fibres are a mixture of different grades that may be reprocessed together. This also explains the deterioration in mechanical properties generally observed. A classification according to CF grade would thus be relevant in order to optimise the reuse of rCFs, as well as a classification according to length, which appears to be necessary. The shorter fibre fractions could be reused in SMCs or BMCs in replacement of vCFs. The longer fibre fractions could be reused in more structural applications. This necessitates a re-alignment of the rCFs, a process which is still under development. Another possibility is reshaping of the rCFs into continuous yarns. Existing spinning techniques have been tested by the University of Leeds and gave promising results, but further work is necessary to improve the resulting yarns. Among all the CF waste in Europe, 60% consists of woven fabrics. It has been shown that they can retain their woven shape after a thermo-chemical treatment, which could be interesting to exploit in terms of structural reinforcements. It might be wise to consider EoL waste separately from production waste, and among the latter cured thermoset from uncured thermoset and dry fibres. Uncured CF materials and dry fibre waste would not need to go through a fibre-matrix separation process. If necessary pre-impregnated fibre waste could be treated in a separation process but under significantly milder conditions. In order to facilitate the reuse of rCFs it

seems to be necessary to develop ready-to-use semi-products, like those using vCFs to manufacture SMCs, BMCs, laminates or preforms. This could be the key to unlocking the reuse of rCFs in real applications. Specific characterisation standards are also required to give a framework and reference points to potential reusers. All these efforts are necessary to encourage widespread industrial and customer acceptance.

It was suggested in the literature that vGFs could be replaced by rCFs, however LCA studies showed that this was not economically viable as rCFs are more expensive than vGFs. Furthermore, the global environmental impact is also worse. Nevertheless, it was confirmed that benefits to the environment can arise from the replacement of vCFs by rCFs, mainly thanks to the energy saved by avoiding the production of CFs.

The organic fractions recovered after thermo-chemical separation have received little attention. It has been stated that this is not economically viable, but in spite of this valuable products have been identified after resin degradation and successfully reused in new resins. Considering that resin prices follow the evolution of the petroleum prices, it might be of interest to develop viable methods to recover monomers from resins. In a similar light, recyclable resins have begun to be developed. These could judiciously be used to manufacture composites with recycled fibres, so that it would be unnecessary to go through the same reclamation processes several times. Indeed, we may assume that the number of recycling treatments gradually affects the reinforcement properties of the fibres, at least with regard to the required size-reduction post-treatment.

Highly structural applications are unlikely to incorporate recycled fibres, in particular those made from long CFs; new fibres will always be necessary. Efforts are also necessary in order to produce cheaper carbon fibres.

The recycling of composite materials is on the right track, but challenges still have to be taken-up in order to finally make it a commercial reality. The innovation has just started in this field and for this reason it is also a source of opportunities.

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