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Recycling of carbon fiber reinforced composites waste to close their Life Cycle in a Cradle-to-Cradle approach.

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Carbon fibers, Composite Materials, Recycling, Process monitoring and control, Pyrolysis, Thermal recycling, Chemical recycling, Solvolysis, Fiber/matrix bond, LCA, Biobased polymer, Sustainability, Circular economy.

GRAPHICAL ABSTRACT

Only for TOC

Recycling of carbon fiber reinforced composites waste to close their Life Cycle in a Cradle-to-Cradle approach.

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Abstract

Carbon fibre reinforced polymers (CFRPs), with a demand expected to reach 194 ktons by 2022 and a global market increase to $48.7 billion, are increasingly popular materials due to their ability to conjugate superior mechanical resistance and lightness, thus allowing their widespread ranging from aerospace and wind turbines to automotive and sporting goods. A foreseeable consequence is the growth of production scraps and end–of–life (Eol) composites. Considering the still high cost of the virgin CF and a CF demand expected to reach 117 ktons by 2022 (average of 30 €/kg and energetic cost of 183–286 MJ/kg), this review outlines recent advances of the existing methods to recycle cumulative composite wastes, still with many unresolved problems and issues, with emphasis on CF recovery and understanding their retained properties. Finally, a brief overview on the companies that offer CFRPs recovery services with the aim of addressing the issue of Eol is presented.

The global market for carbon fiber composites and opportunities

In recent decades, carbon fibers (CF) have found widespread application in a growing number of fields, such as automotive, aerospace and defense, sea-vehicles, wind turbines, storage tanks, sport, and leisure [1-3]. Their utilization as high-performance light-weight reinforcement has just recently had a boost mainly in high added value applications. The CF industry has been steadily growing, and lately spreading towards more mass-oriented market segments such as the mainstream automotive and motorcycles, building construction and wind energy, where they are applied in the form of CF Reinforced Polymers (CFRP) to replace metal parts in order to provide them with high specific strength and stiffness, lighter weight and in turn lower CO_2 emissions.

The analysis of such trends suggests that the world production of carbon fibers, which already almost doubled in the 2009-2014 timespan going from 27 ktons to 53 ktons, will peak at an expected request of 117 ktons by 2022; such exponential progress is estimated to grow annually at 6.6% rate in the value market value, that is expected to reach about $ 12 billion [4], with an obvious parallel expansion of the CFRP segment, which is expected to top a production of about 194 ktons in 2022 [5,6] with a global market increase of about $ 48.7 billion [7].

Such a boost in the CFRP exploitation is now raising the awareness about their fate: a direct consequence of the increased carbon fibers composite production is, indeed, a strong increase in CF-related wastes, coming both from the manufacturing processes (prepreg offcuts; offcuts and scraps of cured composites, which represent about 30-40wt% of the total materials) and, belatedly, from the End of Life products (EoL). Indeed, the global CFRP waste is foreseen to reach up to 20 ktons annually by 2025 [8]. As an example, about 12000 aircrafts worldwide are expected to reach their End-of-Life within the next two decades; however this estimation only slightly comprise dismantling highly CFRP-laden Boeing 787 and Airbus A350 XWB, which are recent model and are expected be in use for 25-30 years-service [9]: indeed a certain delay is expected between the boost in the production and the analogous trend in the waste production. Moreover, it should be also pointed out that current EU legislation, is still lacking a specific regulation for composites’ waste treatment. Some hint is included in the 2000/53/ EC EU Directive, which requires a 95% recovery and 85% recycling extent of total End-of-Life Vehicle weight by 2015 and limits the use of non-metal components if not complying with the Directive requirements, but no specific instruction on how to treat EoL CFRP is specifically addressed

Up to now, incineration and landfilling are the main approaches for disposing of composite wastes (**Figure 1**). These routes, however, are not viable tools in view of the strong expected growth in waste production, since they completely discard the related environmental impact, the waste accumulation of composites and, in particular, they imply the loss of all the CF high added-value [10-16]. It is thus important, starting already from now, to implement different recycling methods for the CFRPs waste [17], to prevent potential issues in the future, that should aim at recovering at least the most precious CF, since CFRP recycling might represent a great resource in the further development of the composite materials [18]. In fact, it has been evaluated that the landfill stocked waste composites worth sums up to €14.7 million of recycled carbon fiber (Re-CF), considering €10/kg as the market price [19], in the case they could be conveniently recovered and recycled. It should be also pointed out that while CFs are the highest added value component in a composite, it is also the most environmentally impacting, due to the fossil raw materials and the strongly energy intensive processing underlying their production.

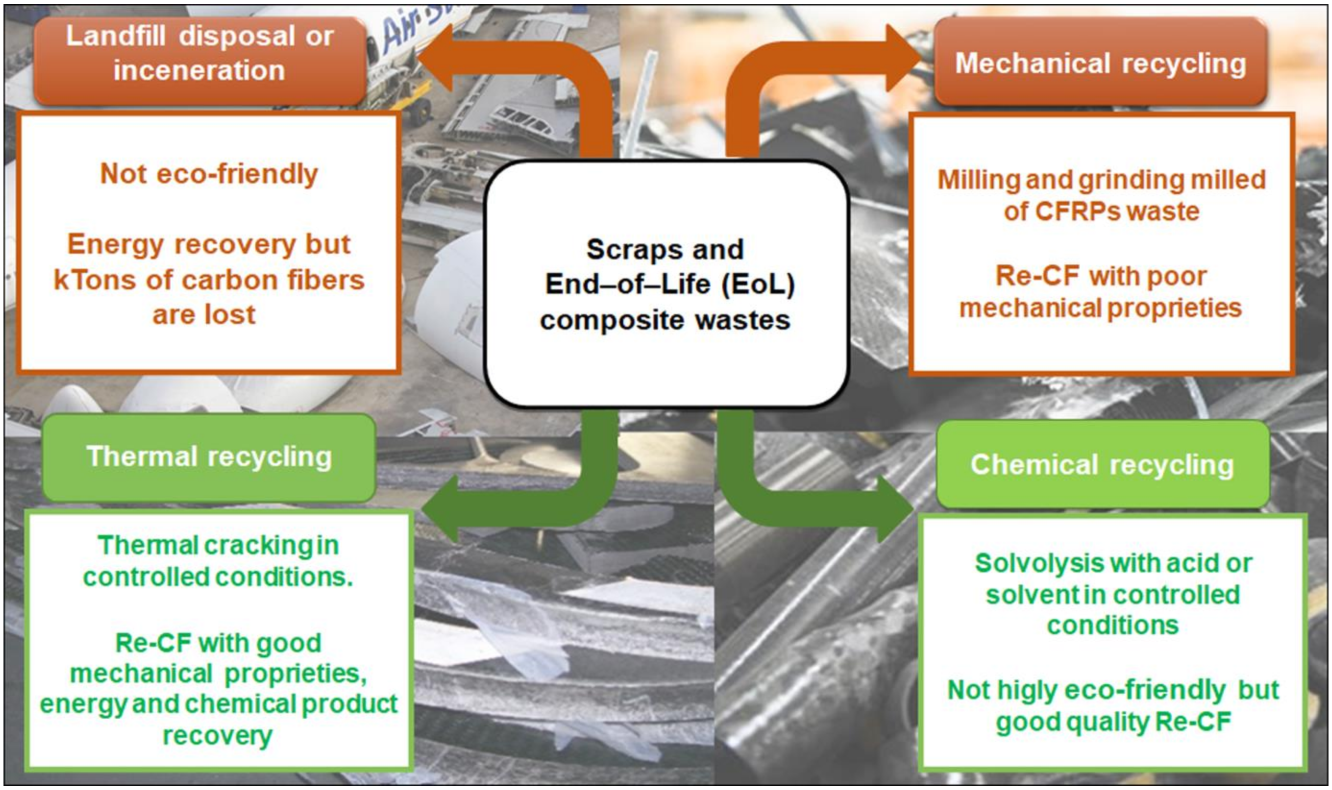

Figure 1. Landfill disposal and the principal advantaged and critical issue of CFRP's recycling methods to Recovery and Reuse of scraps and EoL

Furthermore, while attempts are made at obtaining green composites, intended as more sustainable composites, they often focus just on the study of new, more sustainable matrices. A great deal of literature is devoted to the production of biobased resins [20-24] exploiting biobased resources which, however, being thermosetting, cannot be easily recycled or recovered, neither by disassembling of their components nor by re-melting and remoulding, thus not helping a final recycling process. Another approach involves the switch to biobased thermoplastic materials by green chemistry [25-27], with the production of composites, nanocomposites and green nanocomposites [28-30], that, however, are still far from reaching CFRP performance and a convenient End-of-Life fate.

So while at the present the search for sustainability in composites shows potential for biobased matrices, but no significant improvement in the sustainability of the reinforcement, the research for sound recycling CFRP processes is more and more investigated. Though several recycling techniques are presently available for treating CFRP, they are still far from being optimized and are characterized by some serious drawbacks (Figure 1). This paper aims at reporting the current state of art of CF recycling methods (Figure 2) and to discuss the future perspectives in circular economy prospective.

Recycling methods

The main recycling methods applied to thermosetting CFRP can be classified into three types: (a) mechanical, b) thermal and c) chemical recycling [8,31-34] (**Figure 2**):

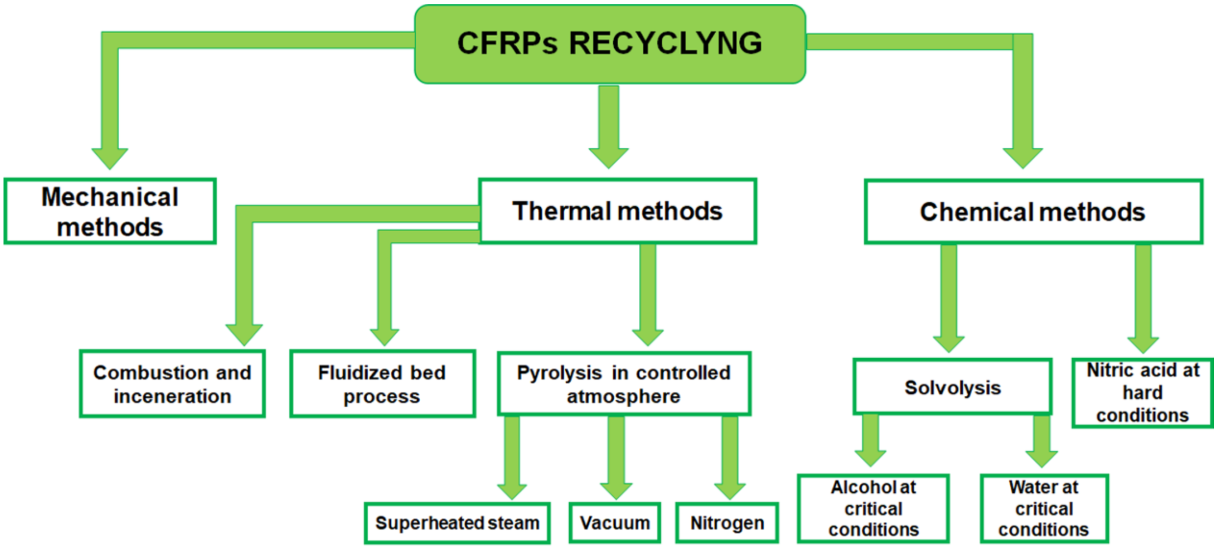

Figure 2. The principal recycling methods of CFRP

Several companies (**Table 1**) offer CFRPs recovery services and their recycling into marketable products [7,34-38]. It is worth noting that pyrolysis process is currently predominately used for CF recovery, as it is well developed, can be applied at different stages of composite manufacturing, and is commercially viable.

Mechanical recycling

Mechanical recycling, consists in first reducing the CFRPs size to 50-100 mm, generally using low speed cutting or crushing mills; then wastes are milled or grinded at high speed to reach about 50 μm (powder) or 10 mm (fibrous) size [31]: this process requires a significant energy consumption since shredding fibers is not easily attained. In this way, moreover, the structural integrity of Re-CF is not preserved, leading to a significant loss of the mechanical properties and, above all, in the economic value [39]. A different approach to attain mechanical recycling is the electrodynamic fragmentation (EDF): CFRPs waste is shredded by means of a high voltage impulse (between 50 and 200 kV) in ionized water [40,41]. In both cases, the actual field of applications of mechanically recycled Re-CF strongly depends on the obtained particle size [32,42]: however, all the obtained Re-CFare mainly used in low-value applications, chiefly as fillers or particle reinforcements [32].Mechanical recycling can be also applied a pre-treatment step for thermal and chemical recycling , when the waste's original shape and size do not fit process requirements[43,44].

Table 1. Current CFRP composite recycling companies and their technologies

Company	**Technology**	Capacity (tons/year)
Alpha Recyclage Composites (France)	Steam thermolysis process	300
Carbon Conversions Inc. (Toyota Tsusho America, US)	Pyrolysis	2000
CFK Valley Stade Recycling GmbH & Co. KG (Germany)	Pyrolysis	1000
Curti SpA (Italy)	Pyrolysis	120
ELG Carbon Fibre (UK)	Pyrolysis	2000

Hitachi Chemical	Solvolysis	12
KARBOREK RCF (Italy)	Pyrolysis	1000
Procotex (Belgium)	Mechanical (Pulling, milling and precision cutting to length	N/A
SGL Automotive Carbon Fibres (US)	Pyrolysis	1500
Takayasu	Pyrolysis	60
Toray Industries	Pyrolysis	1000
University of Manchester (UK)	Mechanical	20
University of Nottingham (UK)	Fluidized bed	100
V-Carbon (US)	Solvolysis	1.7

Thermal recycling

Thermal recycling is subdivided into three types [32], however the underlying principle remains the same, that is the use of high temperatures for degrading the polymer matrix to leave the fibers as a residue (**Figure 2**). Thermal treatments need thorough control of the process parameters (atmosphere, temperature and residential time) in order to avoid loss of valuable products or undesired modifications in the chemistry of the recovered fractions [33,45,46]. When thermal recycling is carried out solely for energy recovery, as in combustion and incineration treatments, the process leads to the loss of valuable materials (CF), as well as to the production of polluting emissions, imposing the use of expensive gas cleaning devices, and of large quantities of ashes, which are considered an inert waste that has to be nonetheless disposed of. Contrarily, in the case of pyrolysis and fluid-bed processes [47], it is possible to obtain clean recycled CF as the solid residue of the process. The residential time in the reactor and the process temperature (450–700 °C) vary as a function of the polymeric resin to be treated and are of paramount importance for the quality of the recovered residue: indeed, when temperature is too low the fibers surface is covered in an amorphous carbon layer (char) as a consequence of poor matrix degradation: Re-CF are thusstiff, with poor mechanical properties and scarce interface interactions when reimpregnated. When, instead, the temperature is too high, CF's surface can partially oxidized, with a consequent reduction in the fibres' diameter and, in turnin their mechanical properties [30,48,49].

Pickering and his research group started developing the fluidized bed recycling since the 2000s [45] and this technology is now effectively operational at a pilot-scale stage to treat also CFRPs. The process requires shredding the parts to typically 6~25 mm, then the composite waste is fed into a bed of silica sand (with size around 0.85 mm) at about 450-550 °C under a hot air flow (0.4-1.0 m /s). The composite waste is separated into fibers and volatile compounds. The latter are removed from the air flow and allowed to pass into a second oxidation chamber at 1000°C [45]. This process produces non-oriented Re-CF with a length between 5-10mm; it is also characterized by a low energy consumption with respect to production of virgin fibers [50]. However, the obtained Re-CF show only a 10-75% retention of the pristine tensile strength, which significantly limits their reuse.

The other relevant thermal approach is represented by pyrolysis (**Figure 3**), which appears also to be the most appealing, a process in which organic materials are thermally decomposed into simpler components when subjected to strong heat (450-700°C) under an oxygen deprived atmosphere. When pyrolysis is applied to CFRPs, leads to the thermal cracking of the matrix fraction, no matter if thermoplastic or thermosetting, producing volatiles that flow away from the reactor and can be subsequently separated into two portions: a condensable component (pyrolysis oil) and a non-condensable fraction (gas). Both these fractions can be used either as a source of valuable feedstock for further manufacturing chemicals other than from fossil resources[51] or as fuel. The high carbon and hydrogen content of both volatile and non-volatile components result in a high calorific value which can be profitably used to practically fully sustain the pyrolysis process itself [49]. At the end of the process a solid residue of CF can be recovered. Reports from industry suggest that the production of Re-CF through pyrolysis of CFRPs waste will consume only 5–10% of the energy required for production of virgin fibers [33,34]. During the process, a layer of pyrolytic carbon can form onto the fibers [33,52], which can be removed by additional processing in oxidative conditions [51,53], giving back fibers in a suitable condition to be used as feedstocks in a secondary raw material generation approach with good fiber/matrix adhesion [48,53].

Danish's company ReFiber is recycling aircraft CFRP component waste, making use ofan optimized semi-industrial pyrolysis plant, that has already implemented a secondary heating system to positively eliminate the residue char. To completely avoid such char formation on the Re-CF, ELG Carbon Fibre (UK) , uses a commercial-scale semi-open continuous belt furnace with a controlled atmosphere [33,54], CFK Valley Stade Recycling GmbH (Germany), and Materials Innovation Technologies–Reengineered Carbon Fiber (MIT-RCF) (US), instead use an industrial continuous pyrolysis process: the large furnace and continuous flow allow them to recover longer and cleaner Re-CFs [33]. In order to optimize the whole pyrolytic process, Curti SpA (Italy) recently introduced an innovative static-bed batch pilot reactor [55,56] which soon afterwards was modified into continuous process in two steps able to combine at 500-550°C both the pyrolysis and the oxidation step, drawing the main advantages of the different disposal techniques (**Figure 3**).

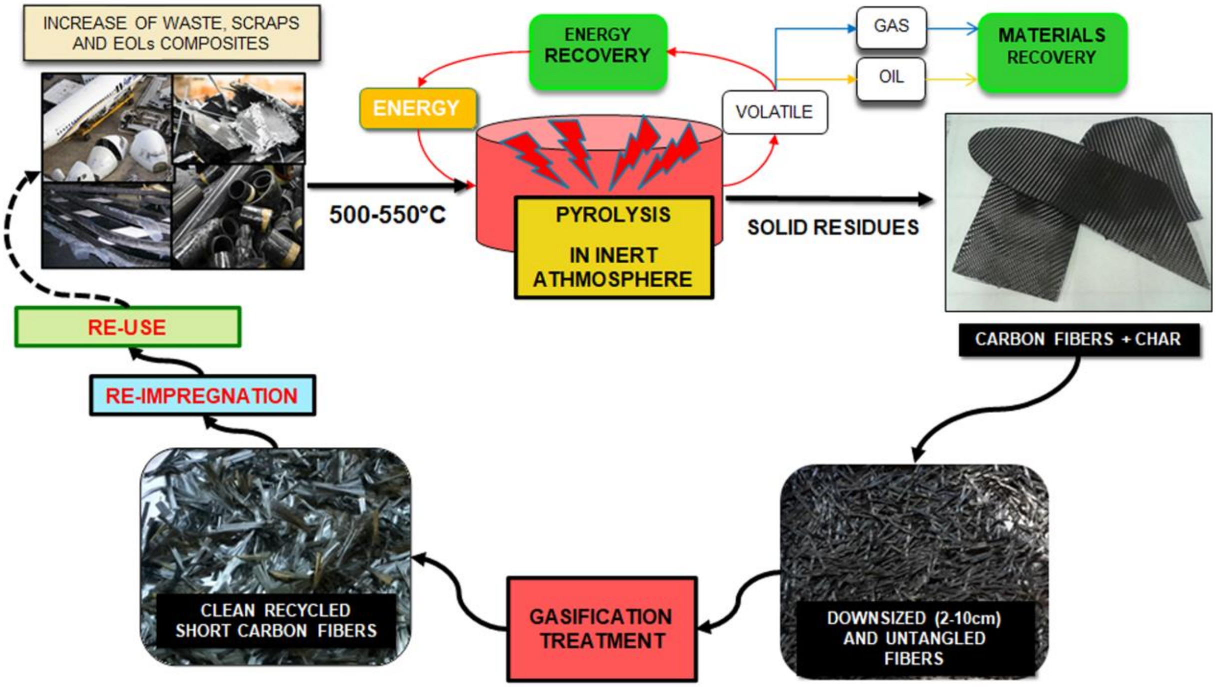

Figure 3. Thermal recovery method in two steps (pyrolysis and gasification) and reuse of CF.

In such a plant, pyrolysis can be carried out on the whole parts, up to 2m in diameter, in order to save the energy costs of shredding the feed wastes, and simultaneously recovering energy and materials with Re-CF retaining 95% of their original tensile strength [57,58]. In these conditions the obtained carbon fibers maintain the original arrangement they had in the waste part, as depicted in Figure 3.

A novel super-heated-steam based method (at 550 °C) has been also recently used to obtain high-quality Re-CF with almost no char residue [59]. Different reaction conditions have been also tested such as the use of CO_2 and water vapor to promote an efficient and effective char removal [60].

Chemical recycling

In chemical recycling process (typically called solvolysis), the polymer matrix is decomposed by a solution of acids, bases and solvents whose composition needs to be tuned on the matrix [33,61] (**Figure 2**). In order to increase the surface area in contact with the solution and promote matrix dissolution, solid CFRP are first shredded; at the end of the process, the Re-CF are washed to remove decomposed polymeric compounds and solvent residues [61,62]. The obtained Re-CF can be long and they showed to retain their tensile strength, with few percentage points drop compared to virgin CFs [63.64]. The use of dangerous and concentrated chemicals has, however, a recognized significant environmental impact [33]. Nitric acid allows for a decomposition of epoxy resins and a better recycling of CF compared to both sulfuric and hydrochloric acid [65,66]. Using ultrasonic solvolysis in diluted nitric acid and H_2O_2 at a temperature below 60°C it is also possible to reach a matrix's decomposition extent of 95% with high efficiency [67]. The use of acetone+H_2O_2 [67], DMF+H_2O_2 [68] and an aqueous mixture of peracetic acid allowed to obtain Re-CF and a matrix decomposition extent ranging from 90 to 97% [68-70]. The solvolysis can be also carried out in supercritical or subcritical condition using nontoxic water and alcohol solvents with critical pressure and temperature conditions [71-73]. However, the process is not yet commercialized, and the operating conditions require higher energy consumption with respect to traditional chemical recycling.

Future challenges

It is a fact that, when taking into account factors such as climate change, global warming, environmental sustainability and circular economy, the landfill or incineration of CFRP wastes must be avoided. In the last two decades, several technologies for CFRPs recycling (mechanical, thermal, and chemical) have been implemented, especially in Europe and US, and new technologies are more and more sought after. In this context, more efforts are required to improve the technology readiness level (TRL) of the processes discussed in this paper and their scalability should be economically accessed. The pyrolysis process, with an actual capacity of about 7 ktons/y respect to production of CFRPs waste of about 170 ktons/y, was identified as the most viable and sustainable CFRP recycling process to achieve process and resource efficiency. This process leads to CF recycling together with the recovery of gaseous and liquid products obtained by matrix degradation, which can be further used as raw materials.

The current state of art states that for several applications it is possible to replac virgin CF with Re-CF (TNT, SMC and BMC technologies) [6,9,18,19,50,53].

To develop commercially viable recycling activities, the future researches must be focused on the following points: achievement of consistent quality of recycled fibers; reuse of Re-CF as reinforcement in thermosetting and moistly thermoplastic polymers, also from renewables sources; study of mechanical properties after reuse and of remanufactured technologies; evaluation of the potential to close the life-cycle loop f CFRPs; reducing energy consumption and recycling cost with a potential effective target of 5 €/kg. Furthermore, the principal challenging issue is creating new opportunities and applications for expanding the Re-CF use in other commercial fields at high market value that can compete with metallic counterpart.

Conflict of interest statement

Authors declare no conflict of interests.

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