Recycling of carbon fiber reinforced composite waste to close their life cycle in a cradle-to-cradle approach

Published Version:
Giorgini L., Benelli T., Brancolini G., Mazzocchetti L. (2020). Recycling of carbon fiber reinforced composite waste to close their life cycle in a cradle-to-cradle approach. CURRENT OPINION IN GREEN AND SUSTAINABLE CHEMISTRY, 26, 1-8 [10.1016/j.cogsc.2020.100368].

Availability:
This version is available at: https://hdl.handle.net/11585/769126 since: 2024-05-31

Published:
DOI: http://doi.org/10.1016/j.cogsc.2020.100368

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

Loris Giorgini[1,2,*], Tiziana Benelli[1,2], Gianluca Brancolini[1], Laura Mazzocchetti[1,2]

[1] *Department of Industrial Chemistry "Toso Montanari", University of Bologna, Viale Risorgimento 4, I-40136 Bologna, Italy*

[2] *Interdepartmental Centre for Industrial Research – Advanced Mechanics and Materials, University of Bologna, Viale Risorgimento 2, I-40136 Bologna, Italy*

**Corresponding author:*

Tel: +39-051-2093688; Fax: +39-051-2093651

E-mail: loris.giorgini@unibo.it (*Loris Giorgini)*

Keywords

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.

Loris Giorgini[1,2,*], Tiziana Benelli[1,2], Gianluca Brancolini[1], Laura Mazzocchetti[1,2]

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.

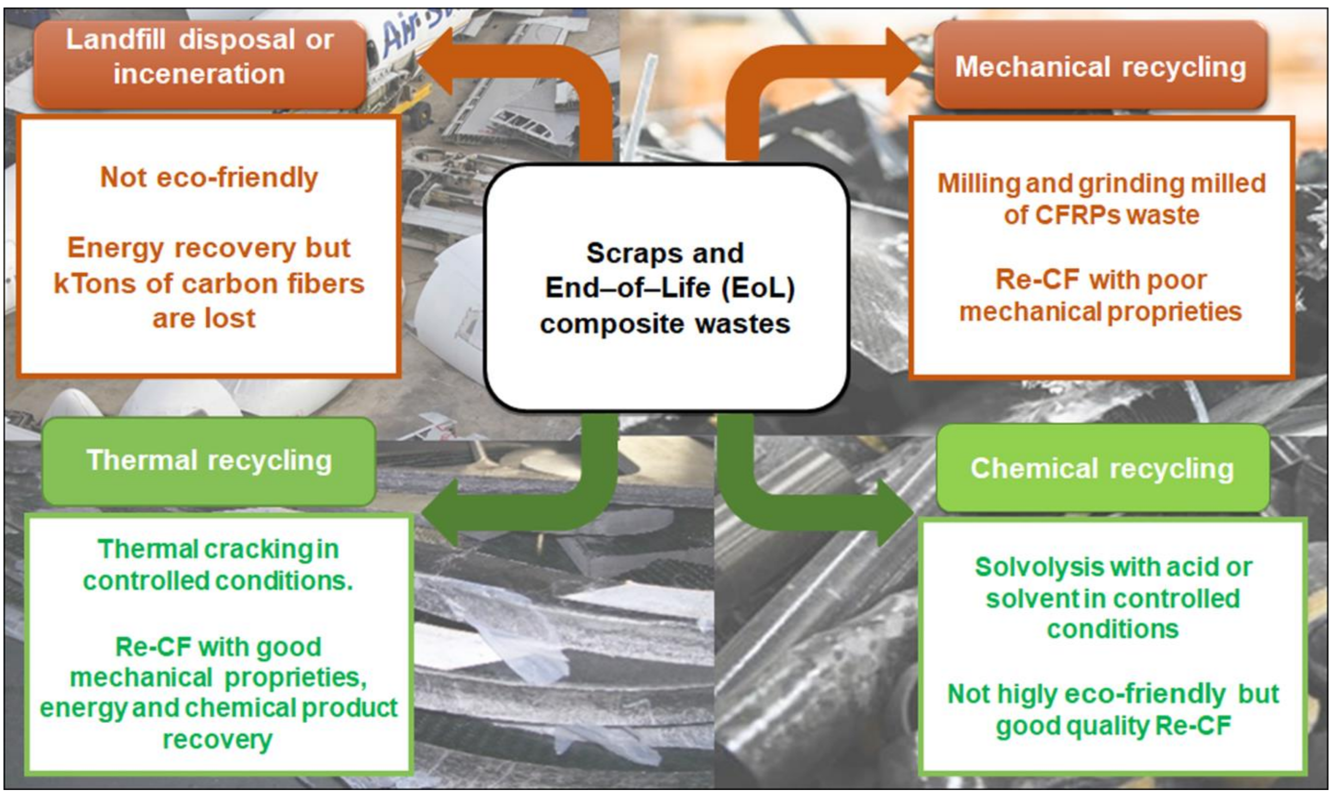

Figure 1. Landfill disposal and the principal advantaged and critical issue of CFRP's recycling methods to Recovery and Reuse of scraps and EoL

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**):

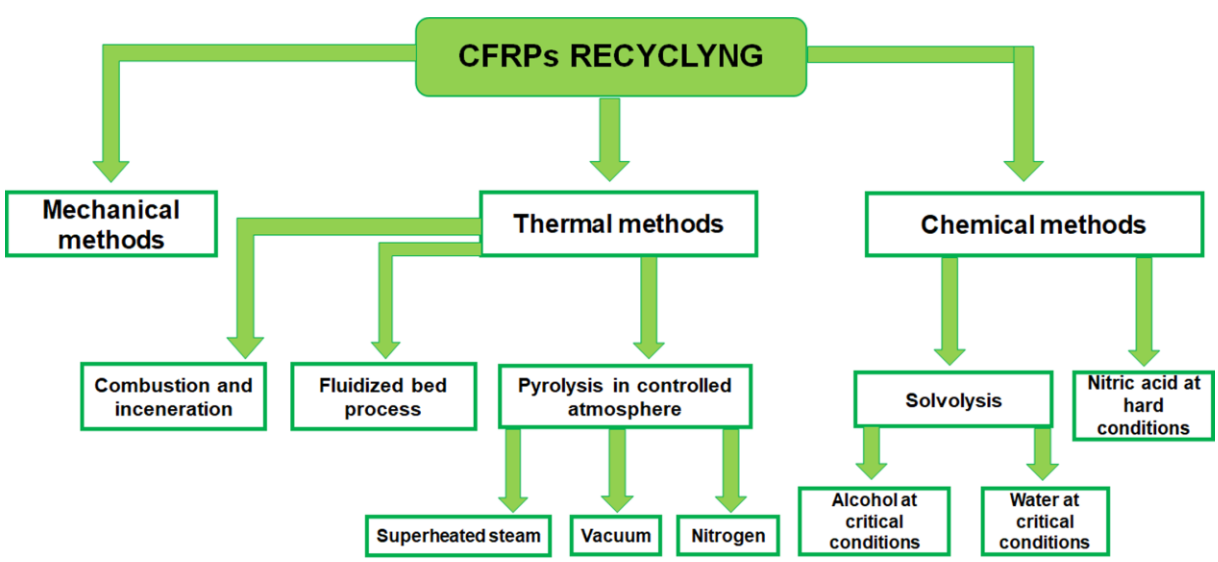

Figure 2. The principal recycling methods of CFRP

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**).

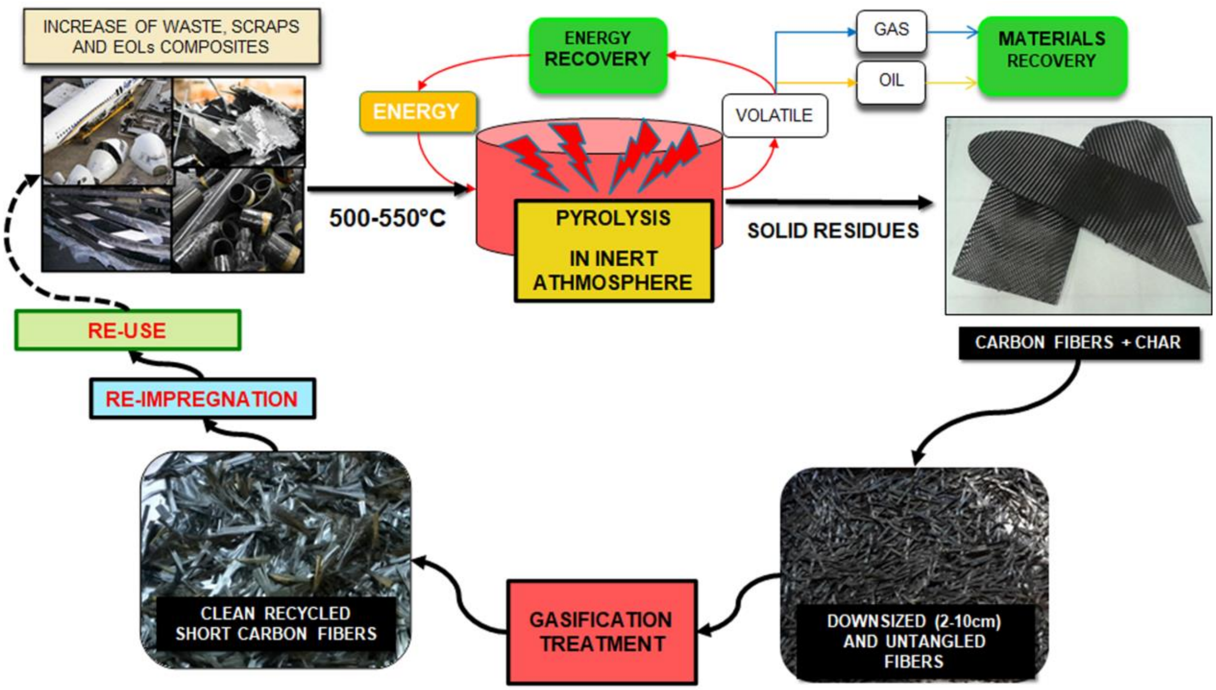

Figure 3. Thermal recovery method in two steps (pyrolysis and gasification) and reuse of CF.

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.

Acknowledgement

This publication was supported by the Project TEAM SAVE, code E91B18000460007, funded within POR FESR 2014-2020 action by Regione Emilia Romagna.

Papers of particular interest, published within the period of review, have been highlighted as:

* of special interest

** of outstanding interest

References.

1. Chung DDL: **Processing-structure-property relationships of continuous carbon fiber polymer-matrix composites.** *Mat Sci Eng R* 2017, **113**:1–29.

2. **Composite Materials Market – Forecast (2020 - 2025).** IndustryARC, 2020, https://www.industryarc.com/Report/246/composite-materials-market-analysis-report.html. [Accessed on 9 May 2020]

** A complete and comprehensive report on Composite Materials Market: by material (fibre reinforced polymers, resins, matrices, others), by application (aerospace, construction, wind energy, automotive, others), by method (open moulding & closed moulding) and by geography - forecast

3. Koumoulos EP, Trompeta AF, Santos RM, Martins M, Monterio dos Santos C, Iglesias V, Böhm R, Gong G, Chiminelli A, Verpoest I, Kiekens P, Charitidis CA: **Research and Development in Carbon Fibers and Advanced High-Performance Composites Supply Chain in Europe: A Roadmap for Challenges and the Industrial Uptake.** *J Compos Sci* 2019, **3**:86 1-28.

4. Naqvi SR, Prabhakara HM, Bramer E, Dierkes W, Akkerman R, Brem G: **A critical review on recycling of end-of-life carbon fibre/glass fibre reinforced composites waste using pyrolysis towards a circular economy**. *Resour Conserv Recycl* 2018, **136**:118–129.

** A review on carbon and glass fibers reinforced composites (CFRC/GFRC) recycling via pyrolysis processes and highlight their technical challenges and re-use possibilities in high performance composites.

5. Kraus T, Kühnel M, E Witten: Composites market report 2016 – market developments, trends, outlook and challenges. 2016. https://www.carbon-composites.eu/media/2449/market_report_2016_ccev-avk.pdf. [Accessed on 9 May 2020]

6. Witten E, Mathes V, Sauer M, Kuhnel M: **Composites Market Report 2018-Market developments, trends, outlook and challenges**. 2018. https://www.avk-tv.de/files/20181115_avk_ccev_market_report_2018_final.pdf. [Accessed on 9 May 2020]

7. Meng F, Olivetti EA, Zhao Y, Chang JC, Pickering SJ, McKechnie J: **Comparing Life Cycle Energy and Global Warming Potential of Carbon Fiber Composite Recycling Technologies and Waste Management Options**. *ACS Sustain Chem Eng* 2018, **6**:9854-9865.

8. Zhang J, Chevali VS, Wang H, Wang C-H: **Current status of carbon fibre and carbon fibre composites recycling**. *Compos B. Eng* 2020, **193**:108053.

* A presentation of the current technologies for re-manufacturing CFRPs with recovered carbon fibers.

9. Wong K, Rudd C, Pickering S, Liu XL: **Composites recycling solutions for the aviation industry**. *Sci China Tech Sci* 2017, **60**:1291–1300.

10. Hadi P, Ning C, Ouyang W, Xu M, Lin CS, McKay G: **Toward environmentally benign utilization of on metallic fraction of waste printed circuit boards as modifier and precursor**. *Waste Manag* 2015, **35**:236–246.

11. Liu P, Meng F, Barlow CY: **Wind turbine blade end-of-life options: an eco-audit comparison**. *J Clean Prod* 2019, **212***:*1268–1281.

12. Pillain B, Loubet P, Pestalozzi F, Joerg W, Arnaud E, Cyril A, Guido S: **Positioning supercritical solvolysis among innovative recycling and current waste management scenarios for carbon fiber reinforced plastics thanks to comparative life cycle assessment**. *J Supercrit Fluids* 2019, **154**:104607.

13. Li X, Bai R, McKechnie J: **Environmental and financial performance of mechanical recycling of carbon fibre reinforced polymers and comparison with conventional disposal routes**. *J Clean Prod* 2016, **127**:451–460.

14. Prinçaud M, Aymonier C, Loppinet-Serani A, Perry N, Sonnemann G: **Environmental feasibility of the recycling of carbon fibers from CFRPs by solvolysis using supercritical water**. *ACS Sustain Chem Eng* 2014, **2**:1498–1502.

15. Nunes AO, Viana LR, Guineheuc PM, Aparecida V, Moris S, Faulstich de Pavia JM, Barna R, Soudais Y: **Life cycle assessment of a steam thermolysis process to recover carbon fibers from carbon fiber-reinforced polymer waste**. *Int J Life Cycle Assess* 2018, **23***:*1825–1838.

* A comprehensive Life Cycle Assessment (LCA) approach to recycling CFRP

16. Witik RA, Teuscher R, Michaud V, Ludwig C, Månson JAE: **Carbon fibre reinforced composite waste: an environmental assessment of recycling, energy recovery and landfilling**. *Compos Part A-Appl S.* 2013, **49**:89–99.

17. Moriyama A, Hasegawa T, Nagaya C, Hamada K, Himaki T, Murakami M, Horie M, Takahashi J, Iwahashi H, Moritomi H: **Assessment of harmfulness and biological effect of carbon fiber dust generated during new carbon fiber recycling method**. *J Hazard Mater* 2019, **378**:120777.

18. Roberts T: **Rapid growth forecast for carbon fibre market**. *Reinf Plast* 2007, **51**:10–13.

19. McConnell VP: **Launching the carbon fibre recycling industry**. *Reinf Plast* 2010, **54**:33-37.

20. Mazzocchetti L, Merighi S, Benelli T, Giorgini L: **Evaluation of Tryptophan – Late curing agent systems as hardener for epoxy resin**. *AIP Conf Proceed* 2018, **1981**:020170.

21. Auvergne R, Caillol S, David G, Boutevin B, Pascault JP: **Biobased thermosetting epoxy: Present and future**. *Chem Rev* 2014, **114**:1082-1115.

22. Bobade SK, Paluvai NR, Mohanty S, Nayak SK: **Bio-Based thermosetting Resins for Future Generation: A Review**. *Polym Plast Technol Eng* 2016, **55**:1863–1896.

* Agro-based thermosetting polymers derived from natural resources (cardanol, itaconic acid, tannin, sugar and vegetable oils) are highlighted for future generation with greater sustainability for different applications.

23. Mustapha R, Rahmat AR, Majid RA, Mustapha ANH: **Vegetable oil-based epoxy resins and their composites with bio-based hardener: a short review**. *Polym Plast Technol Eng* 2019, **58**:1311-1326

24. Baroncini EA, Yadav SK, Palmese GR, Stanzione JF: **Recent advances in bio-based epoxy resins and bio-based epoxy curing agents**. *J Appl Polym Sci* 2016, **133**:1-19.

25. Solmi S, Rozhko E, Malmusi A, Tabanelli T, Albonetti S, Basile F, Agnoli S, Cavani F: **The oxidative cleavage of trans-1,2-cyclohexanediol with O2: Catalysis by supported Au nanoparticles**. *Appl Catal A: General* 2018, **557**:89–98.

26. Paone E, Tabanelli T, Mauriello F: **The rise of lignin biorefinery**. *Curr Opin Green Sustain Chem* 2020, **24**:1-6.

27. Tabanelli T, Paone E, Vásquez PB, Pietropaolo R, Cavani F, Mauriello F: **Transfer Hydrogenation of Methyl and Ethyl Levulinate Promoted by a ZrO_2 Catalyst: Comparison of Batch vs Continuous Gas-Flow Conditions**. *ACS Sustain Chem Eng* 2019, **7**:9937−9947.

28. Sisti L, Belcari J, Mazzocchetti L, Totaro G, Vannini M, Giorgini L, Zucchelli A, Celli A: **Multicomponent reinforcing system for poly(butylene succinate): composites containing poly(l-lactide) electrospun mats loaded with graphene**. *Polym Test* 2016, **50:**283–291.

29. Mazzocchetti L, Sandri S, Scandola M, Bergia A, Zuccheri G: **Radiopaque Organic-Inorganic Hybrids Based on Poly (D, L-lactide)**. *Biomacromolecules* 2007, **8**:672-678.

30. Sisti L, Totaro G, Vannini M, Giorgini L, Ligi S, Celli A: **Bio-Based PA11/Graphene Nanocomposites Prepared by In Situ Polymerization**. *J Nanosci Nanotechno* 2018, **18**:1169–1175.

31. Oliveux G, Dandy LO, Leeke GA: **Current status of recycling of fibre reinforced polymers: review of technologies, reuse and resulting properties**. *Prog Mater Sci* 2015, **72**:61–99.

32. Pickering SJ: **Recycling technologies for thermoset composite materials-current status**. *Compos Part A-Appl S.* 2006, **37**:1206–1215.

** A complete and comprehensive first report on recycling technologies for thermosetting composite materials.

33. Pimenta S, Pinho ST: **Recycling carbon fibre reinforced polymers for structural applications: Technology review and market outlook**. *Waste Manage* 2011, **31**:378–92.

34. Gopalraj SK, Kärki T: **A review on the recycling of waste carbon fibre/glass fibre-reinforced composites: fibre recovery, properties and life-cycle analysis**. *SN Appl Sci* 2020, **2**:433.

** Recent developments on the recycling of carbon and glass fibres are presented and discussed.

35. Khurshid MF, Hengstermann M, Hasan MMB, A, Cherif C: **Recent developments in the processing of waste carbon fibre for thermoplastic composites – A review**. *J Compos Mat* 2020, 1-20. https://journals.sagepub.com/doi/10.1177/0021998319886043. [Accessed on 9 May 2020].

* Recent developments in the processing of waste carbon fibre for thermoplastic composites (injection moulding, nonwoven, tape and hybrid yarn spinning technologies).

36. Rybicka J, Tiwari A, Leeke GA: **Technology readiness level assessment of composites recycling technologies**. *J Clean Prod* 2016, **112**:1001-1012.

37. Holmes M: **Recycled carbon fiber composites become a reality.** *Reinf Plast* 2018, **62**:148–153.

38. Amaechi CV, Agbomerie CO, Orok EO, Job S, Ye J: **Economic Aspects of Fiber Reinforced Polymer Composite Recycling**. *Encyclopedia of Renewable and Sustainable Materials* 2020; **2**:377-397.

39. Kaya M: **Recovery of metals and nonmetals from electronic waste by physical and chemical recycling processes**. *Waste Manage* 2016, **57**:64-90.

40. Schinner G, Brandt J, Richter H: **Recycling carbon-fiber-reinforced thermoplastic composites**. *J Thermoplast Compos Mater*, 1996, **9**:239-245.

41. Roux M, Eguémann N, Dransfeld C, Thiebaud F: **Thermoplastic carbon fibre-reinforced polymer recycling with electrodynamical fragmentation: from cradle to cradle**. *J Thermoplast Compos Mater* 2017, **30**:381–403.

42. Palmer J, Ghita OR, Savage L, Evans KE: **Successful closed loop recycling of thermoset composites**. *Compos A. Appl Sci Manuf* 2009, **40**:490–498.

43. Nekouei RK, Pahlevani F, Rajarao R, Golmohammadzadeh R, Sahajwalla V: **Two-step pre-processing enrichment of waste printed circuit boards: mechanical milling and physical separation**. *J Clean Prod* 2018, **184**:1113–1124.

44. Wang H, Zhang G, Hao J, He Y, Zhang T, Yang X: **Morphology, mineralogy, and separation characteristics of nonmetallic fractions from waste printed circuit boards**. *J Clean* Prod 2018, **170**:1501–1507.

45. Pickering SJ, Kelly RM, Kennerley JR, Rudd CD, Fenwick NJ: **A fluidised-bed process for the recovery of glass fibres from scrap thermoset composites**. *Compos Sci Technol* 2000, **60**:509–523.

46. Jiang G, Pickering SJ, Walker GS, Wong KH, Rudd CD: **Surface characterization of carbon fibre recycled using fluidized bed**. *Appl Surf Sci* 2008, **254**:2588–2593.

* A protocol to chemical recycling by fluidized bed process.

47. Vo Dong PA, Azzaro-Pantel C, Cadene AL: **Economic and environmental assessment of recovery and disposal pathways for CFRP waste management**. *Resour Conserv Recycl* 2018, **133**:63–75.

48. Mazzocchetti L, Benelli T, D’Angelo E, Leonardi C, Zattini G, Giorgini L: **Validation of carbon fibers recycling by pyro-gasification: the influence of oxidation conditions to obtain clean fibers and promote fiber/matrix adhesion in epoxy composites**. *Compos A. Appl Sci Manuf* 2018, **112**:504–514.

* One of the first contribution in the thermochemical recycling by continuous pyro-gasification process.

49. Giorgini L, Benelli T, Mazzocchetti L, Leonardi C, Zattini G, Minak G, Dolcini E, Cavazzoni M, Montanari I, Tosi C: **Recovery of carbon fibers from cured and uncured carbon fiber reinforced composites wastes and their use as feedstock for a new composite production.** *Polym Compos* 2015, **36**:1084–1095.

50. Meng F, McKechnie J, Pickering SJ: **An assessment of financial viability of recycled carbon fibre in automotive applications**. *Compos A. Appl Sci Manuf* 2018, **109**:207-220.

51. López FA, Rodríguez O, Alguacil FJ, García-Díaz I, Centeno TA, García-Fierro JL, González C: **Recovery of carbon fibres by the thermolysis and gasification of waste prepreg**. *J Anal Appl Pyrol* 2013, **104**:675–683.

52. Giorgini L, Leonardi C, Mazzocchetti L, Zattini G, Cavazzoni M, Montanari I, Tosi C, Benelli T: **Pyrolysis of fiberglass/polyester composites: recovery and characterization of obtained products**. *FME Trans* 2016, **44**:405–414.

53. Meyer LO, Schulte K, Grove-Nielsen E: **CFRP-recycling following a pyrolysis route: process optimization and potentials**. *J Compos Mater* 2009, **43**:1121–1132.

* In this study different process parameters during pyrolysis were investigated and optimized in order to provide the reclaimed carbon fibers with properties close to new fibers.

54. Onwudili JA, Miskolczi N, Nagy T Lipoczi G: **Recovery of glass fibre and carbon fibres from reinforced thermosets by batch pyrolysis and investigation of fibre re-using as reinforcement in LDPE matrix**. *Compos B. Eng* 2016, **91**:154–161.

55. Giorgini L, Benelli T, Leonardi C, Mazzocchetti L, Zattini G, Cavazzoni M, Montanari, I, Tosi C: **Efficient recovery of non-shredded tires via pyrolysis in an innovative pilot plant**. *Environ Eng Manag J* 2015, **14(7)**:1611–22.

56. Neri E, Berti B, Passarini F, Vassura I, Giorgini L, Zattini G, Tosi C, Cavazzoni M: **Application of LCA methodology in the assessment of a pyrolysis process for tyres recycling.** *Environ Eng Manag J* 2018, **17**:2437-2445.

57. Mazzocchetti L, Benelli T, Zattini G, Maccaferri E, Brancolini G, Giorgini L: **Evaluation of carbon fibers structure and morphology after their recycling via pyro-gassification of CFRPs**. *AIP Conf Proc* 2019, **2196**:020036.

58. Zattini G, Mazzocchetti L, Benelli T, Maccaferri E, Brancolini G, Giorgini L: **Mechanical Properties and Fracture Surface Analysis of Vinyl Ester Resins Reinforced with Recycled Carbon Fibres**. *Key Eng Mater* 2020, **827**:110-115.

59. Kim KW, Lee HM, An JH, Chung DC, An KH, Kim BJ: **Recycling and characterization of carbon fibers from carbon fiber reinforced epoxy matrix composites by a novel super-heated-steam method**. *J Environ Manage* 2017, **203**:872-879.

60. Limburg M, Stockschläder J, Quicker P: **Thermal treatment of carbon fibre reinforced polymers (Part 1: recycling)**. *Waste Manage Res* 2019, **37**:73-82.

* Carbon dioxide and water vapour were used to remove the carbon residues on the recovered carbon fibers by pyrolysis.

61. Jody BJ, Pomykala JA Jr, Daniels EJ, Greminger JL: **A process to recover carbon fibers from polymer-matrix composites in end-of-life vehicles.** *JOM* 2004, **56**:43-47.

62. Asmatulu E, Twomey J, Overcash M: **Recycling of fiber reinforced composites and direct structural composite recycling concept**. *J Compos Mater* 2014, **48**:593–608.

63. Lee CK, Kim YK, Pruitichaiwiboon P, Kim JS, Lee KM, Ju CS: **Assessing environmentally friendly recycling methods for composite bodies of railway rolling stock using life-cycle analysis**. *Transp Res D Transp Environ* 2010, **15**:197–203.

64. Lee SH, Choi HO, Kim JS, Lee CK, Kim YK, Ju CS: **Circulating flow reactor for recycling of carbon fiber from carbon fiber reinforced epoxy composite**. *Korean J Chem Eng* 2011, **28**:449–454.

65. Meng F, McKechnie J, Turner T, Wong KH, Pickering SJ: **Environmental Aspects of Use of Recycled Carbon Fiber Composites in Automotive Applications**. *Environ Sci Technol* 2017, **51**:12727-12736.

66. Ma JH, Wang XB, Li B, Huang LN: **Investigation on recycling technology of carbon fiber reinforced epoxy resin cured with amine**. *Adv Mat Res* 2009, **79-82**:409–412.

67. Das M, Varughese S: **A novel sono-chemical approach for enhanced recovery of carbon fiber from CFRP waste using mild acid-peroxide mixture**. *ACS Sustain Chem Eng* 2016, **4**:2080–2087.

68. Xu P, Li J, Ding J: **Chemical recycling of carbon fibre/ epoxy composites in a mixed solution of peroxide hydrogen and N, N-dimethylformamide**. *Compos Sci Technol* 2013, **82**:54–59.

69. Li J, Xu PL, Zhu YK, Ding JP, Xue LL, Wang YZ: **A promising strategy for chemical recycling of carbon fiber/thermoset composites: self-accelerating decomposition in a mild oxidative system**. *Green Chem* 2012, **14**:3260-3263.

70. Das M, Chacko R, Varughese S: **An efficient method of recycling of CFRP waste using peracetic acid**. *ACS Sustain Chem* Eng 2018, **6**:1564–1571.

* A protocol to chemical recycling by solvolysis process.

71. Morin C, Loppinet-Serani A, Cansell F, Aymonier C: **Near- and supercritical solvolysis of carbon fibre reinforced polymers (CFRPs) for recycling carbon fibers as a valuable resource: state of the art.** *J Supercrit Fluids* 2012, **66**:232–240.

72. Jiang G, Pickering SJ, Lester EH, Warrior NA: **Decomposition of epoxy resin in supercritical isopropanol**. *Ind Eng Chem Res* 2010, **49**:4535-4541.

73. Piñero-Hernanz R, García-Serna J, Dodds C, Hyde J, Cocero MJ, Poliakoff M, Kingman S, Lester E: **Chemical recycling of carbon fibre composites using alcohols under subcritical and supercritical conditions**. *J Supercrit Fluids* 2008, **46**:83–92.