Journal of Cleaner Production 304 (2021) 127158

Contents lists available at ScienceDirect

Journal of Cleaner Production

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

LCA and LCC of a chemical recycling process of waste CF-thermoset composites for the production of novel CF-thermoplastic composites. Open loop and closed loop scenarios

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ARTICLE INFO

Article history:
Received 10 December 2020
Received in revised form
12 April 2021
Accepted 15 April 2021
Available online 20 April 2021

Handling editor: Cecilia Maria Villas Bôas de Almeida

Keywords:
Circular economy
Polymer waste recycling
Eco-design
Epoxy-thermoplastic
Injection moulding

ABSTRACT

The case study presented in this paper is a follow up of a topic already examined in previous studies relating the life cycle assessment (LCA) of a chemical treatment process used to recycle a specific type of carbon fiber (CF) reinforced thermoset composite. In the present study the LCA is coupled with the life cycle cost (LCC) analysis for the economic assessment. Furthermore, the research sought to specify the best available technology for the reuse of the materials recovered through the chemical recycling process. The new LCA results are more reliable and more current than the scenario presented in the previous LCA studies. In the previous scenario the possibility to recover long carbon fibers “ready to use” was considered. This scenario, even if under investigation by the recycling company, is still not possible for technological limitations as the fibers recovered after the chemical process require further treatments before being used in thermoset composite. Consequently, a more feasible technology was investigated and, according to our laboratory research results, one practical way to recycle the CF-thermoset composites is to shred them before the chemical treatment in order to recover shredded CFs and epoxy thermoplastic from cleavable thermosets. These materials can be easily compounded together to manufacture a CF-thermoplastic composite through injection moulding as we demonstrated herein through some laboratory experiments. The LCA and LCC were accounted for the recycling process via solvolysis up to the recovery phase of the epoxy-thermoplastic resin and the short carbon fibers. The paper presents laboratory test results of the remanufacture of the two reclaimed materials for the production of a thermoplastic CF-composite.

1. Introduction

1.1. Recycling processes of carbon fibers reinforced thermoset composites

The increasing use of carbon fibers reinforced polymers (CFRP) generates an increasing amount of waste, comprising end-of-life (EOL) prepregs, manufacturing cut-offs, testing materials, production tools and EOL components (Pimenta et al., 2011). Consequently, turning CFRP waste into a valuable resource and closing the loop in the CFRP life cycle is vital for the continued use of the material in some applications (e.g. the automotive industry). For this reason, CFRP recycling has received great attention over the last 15 years, not only from researchers but also from several collaborative industrial entities (e.g. Aircraft Fleet Recycling Association [AFRA] and European Composite Recycling Services Company [ECRC]). Full recycling of the composite materials requires reclaiming fibers and matrices which are suitable for remanufacturing in new composites. There are several methods for recycling CFRP and the processes can be classified, depending on the main method used to break down the waste, as: mechanical process, pyrolysis, fluidized bed process and chemical process (Pastine, 2013). Mechanical recycling involves breaking down the composite by, for instance, shredding, crushing or milling. The resulting

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https://doi.org/10.1016/j.jclepro.2021.127158
0959-6526/

reclaimed scrap pieces can then be segregated by sieving into powdered products (rich in resin) and fibrous products (rich in fibers). Because these are low-value products mainly used as fillers, mechanical recycling is mostly limited and mostly used for glass fiber reinforced polymers (GFRP). Pyrolysis, the thermal decomposition of organic molecules in an inert atmosphere (e.g. nitrogen), is one of the most widespread recycling processes for CFRP. During pyrolysis, the CFRP is heated up in the (nearly) absence of oxygen; the polymeric matrix is volatilized into lower-weight molecules and thus is not reclaimed, whereas the CFs remain inert and are eventually reclaimed (Meyer et al., 2009). Reusing matrix products is not economically viable; nevertheless, the resin calorific energy can be recovered, making the pyrolysis process self-sustained. The reclaimed carbon fibers obtained by pyrolysis have found a certain degree of commercial use as demonstrated by the developments achieved by the company ELG Carbon Fiber Ltd. This company developed several products using a remanufacturing approach focused commingling of short carbon fibers obtained by pyrolysis with virgin thermoplastics to form non-woven mats. Oxidation is another thermal process for CFRP recycling, consisting in combusting the polymeric matrix in a hot and oxygen-rich flow (e.g. air at 450–550 °C). The fluidized bed process (FBP), developed at the University of Nottingham, is the most well-known implementation of such a process (Yip et al., 2002). During recycling, CFRP scrap (reduced to fragments approximately 25 mm large) is fed into a bed of silica on a metallic mesh. As the hot air stream passes through the bed and decomposes the resin, both the oxidized molecules and the fiber filaments are carried up within the air stream, while heavier metallic components sink in the bed. This natural segregation makes the FBP particularly suitable for contaminated EOL components. The fibers are separated from the air stream in a cyclone, and the resin is fully oxidized in an afterburner; energy recovery to feed the process is feasible. Most of the recycling techniques for CFRP can provide short fibers already available in the market. Techniques for the remanufacture of reclaimed carbon fibers (e.g.: HiPerDiF, TuFF, and Rotating Drum) are also available and currently under investigation and development (Zhang et al., 2020).

In a recent review by Giorgini et al. (2020) the actual CFRP recycling technologies have been reviewed and the annual capacity analyzed. ELG with their capacity of 2000 tons/year outperform other approaches. Similarly, other companies like CFK Valley Stade Recycling or Carbon Conversions Inc., drove their attention on the pyrolysis as main technology. However, as stated in the paper this process requires high energy and some refinement to remove the char formed on the fiber's surface to allow the reclaimed product to be remanufactured with good fibre/matrix adhesion.

The technique investigated in the present paper uses the chemical degradation of the composites, by means of chemical dissolution reagents that proved to lead to cleaner and less degraded fibres. The process was developed by Connora® Technologies that uses new formulations of epoxy resin-hardeners that can produce recyclable thermosets after curing reaction with epoxy resins. The recycling process then includes two main steps: one regards the crosslink cleavage in the epoxy by means of a combination of temperature and pH (acidic) required to induce the conversion of epoxy into a thermoplastic polymer that can be easily separated in solution from the fibers thus allowing the reclamation of clean reinforcing fibers; one step regarding the remanufacturing of the reclaimed products into new composites (Banatao et al., 2014). The main advantage given by this technique is the possibility to reach the full reclamation of both components (carbon fibers and the epoxy resin) allowing to use them into the remanufacturing process for the production of new polymer reinforced composites. The present paper covers the LCA analysis of the reclamation process and then, on the reclaimed products, it presents the use as a novel composites obtained by reprocessing the recovered thermoplastic with the reclaimed fibers using standard compounding and injection moulding used for thermoplastic composites.

1.2. Rationale of the paper

The case study presented in this paper is a further study about new formulations of epoxy resin and hardeners developed by Connora® Technologies and provided by R*Concept (Spain) in order to obtain recyclable thermosets which can be transformed into a useable epoxy-thermoplastics. The LCA of the chemical recycling process was already published in previous papers (La Rosa et al., 2016, 2018). Hereafter we include the life cycle cost (LCC) analysis for the economic assessment. Furthermore, the technological coverage is now considered by specifying the best available technology for the reuse of the recovered materials through the chemical recycling process. The present study is an example of how the LCA methodology represents the most comprehensive tool to assist the transition from a linear model to a circular model as it takes into account all stages of the materials lifecycle including materials extraction, manufacture, use and waste management, reuse and recycling. The current throwaway culture is defined as "take, make, disposal" which is based on the linear economy (Heidrich et al., 2016). This linear way of consumption is no longer viable due to the rapid growth of population and economy and requires a move to a circular model (Jahani et al., 2019). The circular economy (CE) in Europe is based on the sustainable development goals (SDG) of the 2030 Agenda (Belaud et al., 2019), and the goal is to increase waste valuation and to develop a CE for recycling and reuse and avoiding the use of virgin materials. To achieve a more efficient waste management system, it is necessary to ensure effective consumption of materials at all stages of the life cycle.

In the LCA method, disposal is considered the end of a product, and recycling/reusing the material is the avoidable burden of the system. However, CE considers recycled material as the starting material of another system that leads to complications in defining the functional unit and the limit of the study system. Two functional units should be considered, namely product 1 (primary product) and product 2 (a product derived from recycled material), and these two products are used at different times. Open loop and closed loop scenarios should be considered.

Finally, we also performed experimental analysis on the compounding of the recycled materials mixed with recycled carbon fibres processed by injection moulding. These experiments served to assess the technical feasibility of the recycled compound in the industrial landscape.

2. The LCA and LCC methodology

2.1. Goal & scope

The LCA methodology is an objective procedure for evaluating energy and environmental loads related to a process or activity, carried out through the identification of the energy and materials used and waste released into the environment. The assessment includes the entire life cycle of the process or activity, from the extraction and treatment of raw materials to manufacturing, transport, distribution, reuse, recycling and final disposal. It was defined in this way for the first time by SETAC (Society of Environmental Toxicology and Chemistry) in 1993, introducing the concept of "life cycle": an analysis "from the cradle to the grave", that is, from the extraction of the necessary raw materials until final disposal (SETAC, 1993).

According to the ISO 14040 series (ISO 14040; ISO14044) an LCA study consists of the following phases (specifically described in the following paragraphs):

1. Definition of the objective and field of application
2. Inventory analysis
3. Impact assessment
4. Interpretation of the life cycle

The product system studied in the present paper is a carbon fiber (CF)-thermoset composite panel that is manufactured and recycled via solvolysis.

The *functional unit*, chosen for the present study is the amount of CF-thermoset composite recycled during a batch of the recycling process, quantified in 35.5 kg.

Conceptually, similarly to the LCA model is the criteria that guides the LCC model (Ristimäki et al., 2013), with the substantial difference that the latter focuses on the analysis of the life cycle of a product from a purely economic point of view. In fact, the LCC can be used in business management to help companies in how to reduce the costs of a product (Hunker et al., 2008) while LCA can help to reduce the environmental burdens.

Conventional LCC consists in the measurement and calculation of all the costs associated with a product during the various phases of its life cycle, following the logical thread of the “cradle to grave” (Rödger et al., 2018). The various cost items are discounted using the interest rate that corresponds to the rate of inflation present at the time of drawing up the model.

The LCC model, as well as the LCA model applied in the present paper was divided into four main phases that refer to the four most important cost items: the acquisition of raw materials; the costs related to production, use of the product and its management; maintenance costs; and finally the costs related to the deconstruction or disposal of the product.

2.2. Considerations

As we analyse the process of recovering and reusing the materials, two allocation procedures are to be taken into consideration: closed-loop and open-loop. The closed-loop refers to product systems where there is no change in the inherent properties of the recycled material. The open-loop is applied when the material is recycled into other product systems and undergoes a change in its properties. In this case, the allocation criteria are based on physical properties, economic value and, finally, the number of subsequent uses of the recycled material. This part will be not included in the present paper but will be considered in future studies.

3. Life cycle inventory analysis (LCI) phase description

LCI is fundamental as it forms the basis for assessing the impact of the life cycle, allowing to quantify the flows into and out of the system boundaries. These flows include the use of resources (raw materials and energy), as well as releases into the air, water and soil associated with the system. During this phase, a list is produced containing the quantities of substances consumed and released into the environment and the quantities of matter and energy used. To conduct this analysis, an iterative procedure is implemented which leads to:

- an increase in system knowledge during the data collection phase;
- identification of new data requirements or limitations;
- a change in the data collection procedures so that the objectives of the study are still met.

The simplified procedure followed for our inventory analysis, according to ISO 14040 (2006a) consists of the following phases:

1. Preparation for data collection, which involves the creation of a flow diagram of specific processes, in order to represent and describe in detail all the process units to be modelled, including their interactions and the list of associated data categories.
2. The collection of data necessary for the inventory for each process unit included in the system boundaries, in order to obtain complete knowledge of all units. Primary data for the recycling process were taken at the company production site; secondary data, were taken from updated literature and from the Ecoinvent databases (Ecoinvent v3.5).

3.1. Life cycle inventory list

The inventory data are listed in Tables 1–5.

To produce the carbon fibres, PAN (polyacrylonitrile fibres) is normally used as precursor. As carbon fibers are not available in the Ecoinvent database, PAN is used (available in the database) with the addition of the electricity required for the production of the fibers (Das, 2011). Since they were manufactured in Germany, the transport distance was considered (Table 1). The matrix used for this application is the Polar Bear epoxy resin cured with the Polar Bear R*ecyclable hardener. This formulation (provided by R*concept) is specifically created to obtain a cradle-to-cradle solution which means to obtain composite products that are fully recyclable. As the R*ecyclable hardner is not included in the Ecoinvent database the diethanolamine data was used (Table 2); the consumables are necessary materials in the composite production process and they are listed in Table 3 according to the Vacuum assisted Resin Transfer Moulding process (VaRTM) (La Rosa et al., 2018).

The inventory data of the chemical recycling process at plant level are listed in the following Tables (1–5). The epoxy thermoplastic is not available in the database. We make the assumption that it can be assimilated to polycarbonate (PC) that is available in the database. This assumption is based on the fact that, after recovery it was analyzed and for the chemical structure and the thermo-mechanical properties it can be assimilated to the PC family (La Rosa et al., 2018).

4. Life cycle impact assessment (LCIA)

The data used for the inventory analysis form the basis for the impact assessment phase which consists of estimating the environmental effects of the products, generated as a result of the consumption of resources and releases in the environment. Selected methods are Cumulative energy demand (CED), CML and Recipe end point, all available in the SimaPro 9 software used for the evaluation (Pré Consultants, 2013, 2016).

4.1. 1 LCIA results and discussion

Fig. 1 shows the network of the chemical recycling process. Each unit process described in each box reports the global warming potential (GWP) results as kg CO_2eq.

The red lines indicate created impacts, as they are associated with the use of chemicals and electricity. The green lines indicate avoided impacts, as they are associated with the recovered materials (CFs and the epoxy-thermoplastic).

As we can see, it is evident that the recycling process has a beneficial (avoided) impact on the environment, considering that the recovered carbon fibers represent almost 100%.

The assumption in this case is that the CFs are recovered ready

Table 1
Inventory data of carbon fibers production.

Output	Quantity
Carbon fibers	1 kg
Input	**Quantity**
Polyacrylonitrile fibres (PAN), from acrylonitrile and methacrylate, prod. mix, PAN w/o additives EU-27 S	1 kg
Electricity, high voltage {DE}| market for | Cut-off, S	459 MJ/kg
Transport, freight, lorry 16–32 metric ton, euro3 {RER}| market for transport, freight, lorry 16–32 metric ton, EURO3 | Cut-off, S	2224 kgkm

Table 2
Inventory data of composite matrix.

Output	Quantity
Matrix	0.257 kg
Input	**Quantity**
Epoxy resin, liquid {RoW}| market for epoxy resin, liquid | Cut-off, S	0.206 kg
Diethanolamine {GLO}| market for | Cut-off, S	0.051 kg
Transport, freight, aircraft {GLO}| market for | Cut-off, S	2700 kgkm

Table 3
Inventory data of consumables.

Output	Quantity
Consumables	24.173 kg
Input	**Quantity**
Nylon 66 granulate (PA 66), production mix, at plant RER	1.228 kg
Polyethylene terephthalate (PET) granulate, production mix, at plant, amorphous RER	2.579 kg
Synthetic rubber {GLO}| market for | Cut-off, S	19.11 kg
Polyamide 6.6 fibres (PA 6.6), from adipic acid and hexamethylene diamine (HMDA), prod. mix, EU-27 S	1.256 kg
Transport, freight, lorry 16–32 metric ton, euro3 {RER}| market for transport, freight, lorry 16–32 metric ton, EURO3 | Cut-off, S	150 tkm

Table 4
Inventory data for the composite panel manufacture.

Output	Quantity
Composite	35.5 kg
Input	**Quantity**
Matrix	12.78 kg
Carbon fibers	22.72 kg
Consumables	24.173 kg
Electricity, high voltage {IT}| market for | Cut-off, S	11.6 kWh

to use as long fibers.

The major impact is due to the production of carbon fibers, with an energetic consumption of almost an order of magnitude larger than other materials.

Another evaluation was performed by using the Cumulative Energy Demand as method (single point), to account the electricity consumption required for the production of the composite panel (Fig. 2).

The CED results confirm that the major impact is created by electricity consumption for the production of carbon fibers. Energy consumption, however, is affected by geographical areas. If the same production processes were carried out in Norway, for example, the energy impacts would be lower due to the use of renewable resources. In Fig. 3 a comparison between the production of 1 MJ of electricity, medium voltage, in Italy (orange) and Norway (green) is shown. The exploitation of non-renewable fossil resources in Italy significantly increases the environmental impact.

An evaluation of the damage assessment is reported in Fig. 4 using the ReCiPe Endpoint method. The damage created during the production phase is to a large extent balanced by the materials recovery during the recycling process, with the assumption that the CFs are recovered at a “ready to use” level.

The most common end-of-life practice for thermoset composites has been landfilling for a long time. The comparison reported in Fig. 5 among of the Connora® recycling scenario and the landfilling scenario, shows environmental benefits to all impact categories. This result was expected and can be considered a further confirmation of the consistency of our study.

Table 5
Inventory data for the recycling process.

Output (avoided products)	Quantity
Carbon fibers	22.49 kg
Polycarbonate {GLO}| market for | Cut-off, S	12.33 kg
Input	**Quantity**
De-ionised water, reverse osmosis, production mix, at plant, from groundwater RER S	380 kg
Acetic acid, without water, in 98% solution state {GLO}| market for | Cut-off, S	18 kg
Sodium hydroxide, without water, in 50% solution state {GLO}| market for | Cut-off, S	12 kg
Electricity, medium voltage {IT}| market for | Cut-off, S	106.6 kWh
Output (waste)	**Quantity**
Wastewater - untreated, organic contaminated EU-27 S	380 kg
Waste plastic, mixture {Europe without Switzerland} | market group for waste plastic, mixture | Cut-off, S	0.45 kg

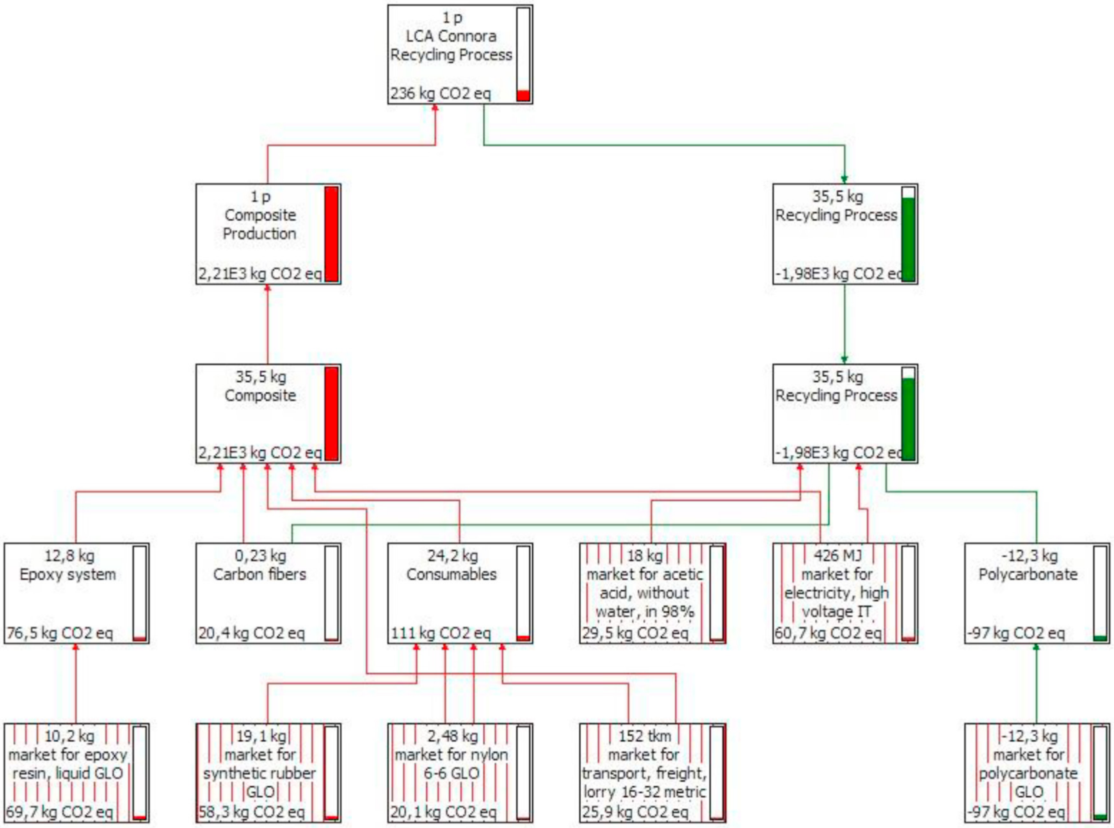

Fig. 1. Network of the recycling process. Scenario of recovering long CFs.

4.2. Sensitivity analysis

Sensitivity analysis was carried out as a procedure to determine how the methodological choices and data affect the results of the LCIA. It is performed as an additional quality analyses of the LCIA (ISO 14044, 2006b) and it is a methodological requirement as life cycle interpretation phase. With the sensitivity analysis it is possible to evaluate the accuracy of the data and their influence on the final results.

One uncertain data in our analysis is the amount of epoxy thermoplastic recovered after the recycling process. We consider the yield of the epoxy-thermoplastic as 100% but it is dependent on the chemical process. Therefore, we conducted a sensitivity analysis on the possibility of change in the yield of the recovered epoxy-thermoplastic. The data set used for the yield are: (100%; 80%; 50%; 40%; 0%). Results are shown in Fig. 6. The impact associated to the recycling process progressively increases proportionally with the increase of the recovered epoxy-thermoplastic yield. This result can be considered as a validation of the applied LCA model.

4.3. LCC results and discussion

For the economic assessment the following flows were considered: materials, energy, transport and labour costs.

The distance between the country of origin of the raw materials

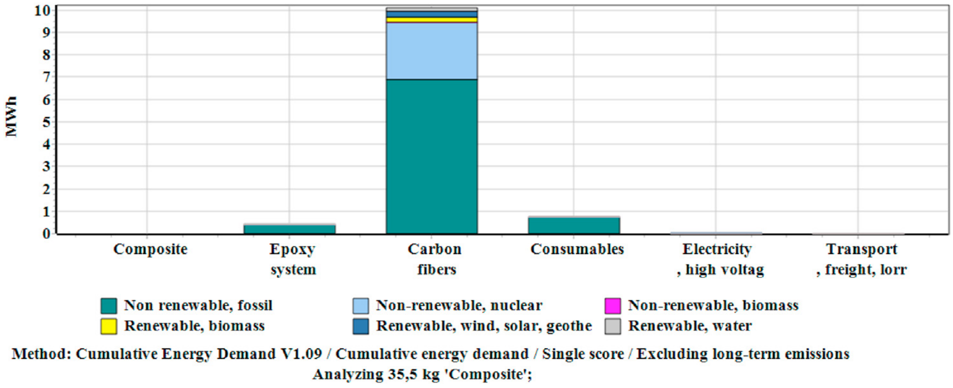

Fig. 2. Electricity consumption for the CF-thermoset panel production.

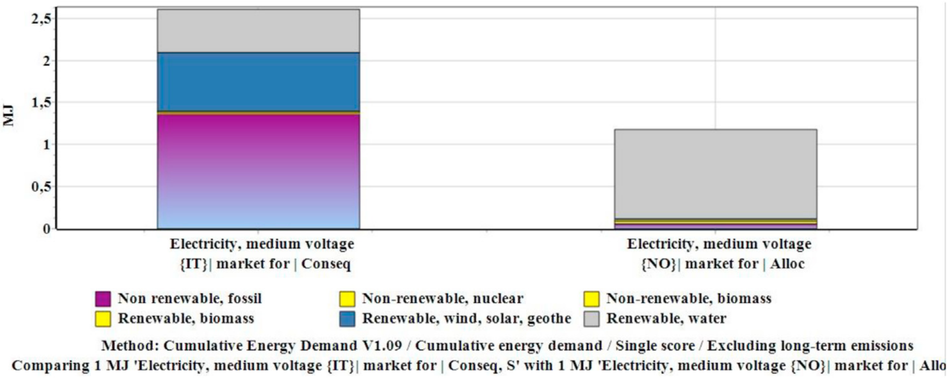

Fig. 3. Country mix production of 1 MJ of electricity in Italy and Norway. CED method.

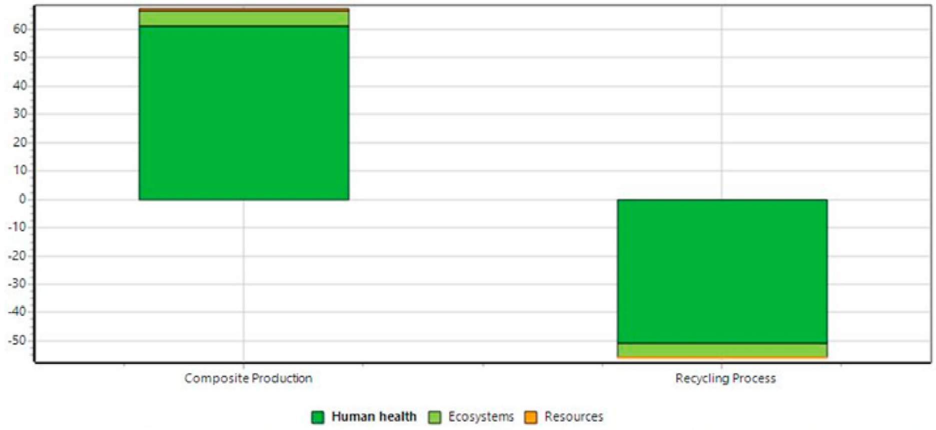

Fig. 4. Damage assessment through ReCiPe Endpoint.

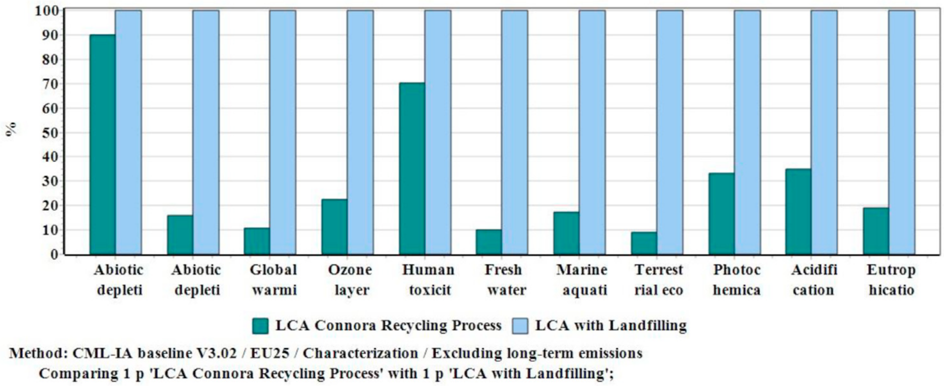

Fig. 5. Comparison of the chemical Connora® recycling scenario and the landfilling scenario.

and the country of production of the composite finite product was accounted in the LCC.

In Table 6 the data inventory for the LCC analysis are reported.

In Fig. 7, the impact evaluation for both production and recycling processes is shown for all LCC impact categories. As we can see, the impact of composite production process is almost completely offset by the avoided impact of the recycling process for energy category, due primarily to the recovering of carbon fibers, as their production requires an intense use of energy.

The costs of all the inputs considered for the LCC are reported in Table 7.

Fig. 8 reports a pie chart of the total cost percentages obtained with the economic assessment method. The cost related to carbon fibers is almost eliminated by the recovered carbon fibers, that are considered ready for the market. Cost of electricity is also reduced from 87.3 € to 2.16 €. These results are obtained in a closed loop system scenario as carbon fibres are assumed to be recycled maintaining the same properties of the virgin fibers and therefore they can go to the market for the same application.

For the cost of the epoxy resin and the hardener a closed loop is

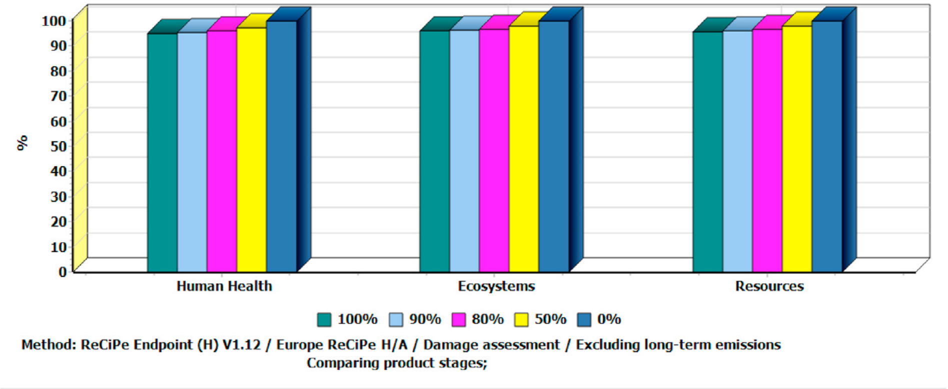

Fig. 6. Sensitivity analysis of the yield of the epoxy-thermoplastic recovered after the chemical recycling process. Data set: 100%; 80%; 50%; 40%; 0% yield.

Table 6
LCC data inventory for materials, energy, transport and labour costs.

Input	Impact category	Factor	Unit
Carbon fiber	Material costs	12.66 (Gill et al., 2016)	€/kg
Consumables	Material costs	5.87	€/kg
Epoxy resin	Material costs	9.95	€/kg
Hardener	Material costs	25	€/kg
Polycarbonate	Material costs	0.84 (Materials and prices, 2020	€/kg
Electricity	Energy costs	0.03	€/kWh
Aircraft fuel	Transport costs	0.0876	€/tkm
Lorry fuel	Transport costs	0.238	€/tkm
Worker	Labour costs	30	€/h

not possible as the recovered materials are chemically different. The avoided impact due to the recovery of the epoxy thermoplastic will be allocated to the following process of reusing the epoxy-thermoplastic in the manufacture a new product.

Efforts are required to optimize the use of consumables as they create large environmental burdens as well as considerably economic costs.

Finally, the economic impact, is affected by geographic areas: the life cost in Norway is shifted to higher values compared to other countries of Europe. The same trend has the labour costs which, compared with Italian ones, are larger. Analyses conducted in different countries will lead, as it should be, to different results.

5. Closed loop and open loop recycling

According to ISO 14044 (2006b), when the recovered materials are used for other applications than the original, or when a material is recycled changing the inherent properties it is an open loop. When a material is recycled without changing the inherent properties it is a closed loop.

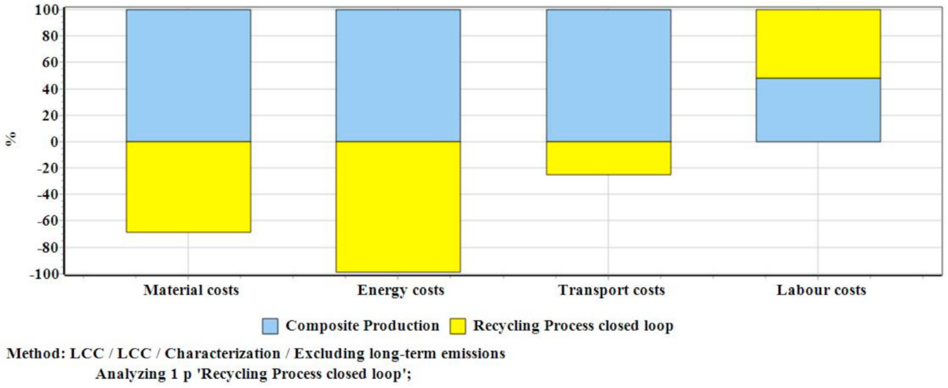

Fig. 7. Economic impact evaluation of composite production and recycling process, closed loop.

Table 7
LCC results for materials, energy, transport and labour costs.

Input	Impact Category	Total cost [€]	Cost for composite production [€]	Cost for recycling process [€]
Carbon fibers	Material costs	2,91	288	−285
Consumables	Material costs	142	142	/
Epoxy resin	Material costs	102	102	/
Hardener	Material costs	63,4	63.4	/
Polycarbonate	Material costs	−10.4	/	−10.4
Electricity	Energy costs	4.43	87.3	−82.8
Aircraft fuel	Transport costs	11.8	11.8	/
Lorry fuel	Transport costs	35.8	47.7	−11.9
Worker	Labour costs	173	82.5	90
Total impact for all categories		524.94	824.7	−300.1

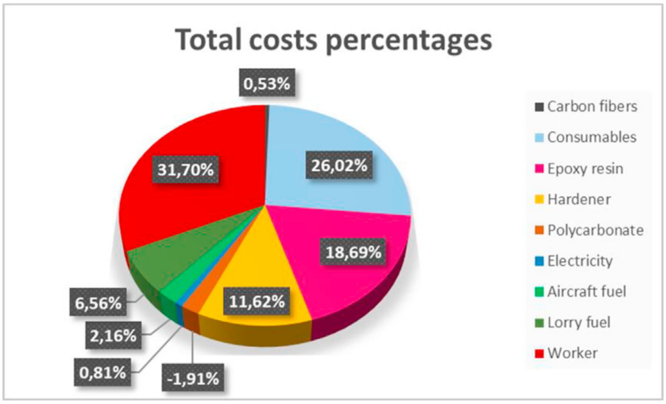

Fig. 8. Pie chart of total costs percentages.

5.1. LCC of the recycling of carbon fibres

The scenario presented so far for the recycled CFs is a closed loop system as they are assumed to maintain the same properties of the virgin carbon fibers. But this scenario has technological limitations due to the fact that the recovery of carbon fibers, in the shape of long fibers, is still not feasible. As results from our laboratory research one practical way to recycle the CF-thermoset composites is to shred them and treat with the chemical solution. In this case the carbon fibers will be recovered as shredded CFs with properties and costs different than the virgin carbon fibers. This is an open loop recycling scenario which means that the avoided impact due to the recovery of the CF are not accounted as avoided impact in this LCA namely we cannot see any benefit in term of cost reduction or environmental burden reduction because they will be accounted in the following process of reusing the recycled CFs to manufacture a new product. Results of the closed loop and open loop scenario for the carbon fibers recycling process are shown in Fig. 9a) and b). Fig. 9 a) reports the LCC of the closed loop system scenario where the recycled carbon fibres are assumed to maintain the same properties and the same costs of the virgin CFs. In this case there is an avoided cost due to the recycled CFs that is accounted as a reduction in the total cost of the CF in the entire life cycle. As result, the cost of the CFs used to produce 35.5 kg of composite is 2.91 €. Fig. 9 b) reports the LCC of the open loop system scenario. The recycled carbon fibers are assumed to have different properties and cost. In this case the avoided costs due to the CFs recovery will be accounted in the LCA of the reuse of these materials and therefore in this scenario the cost of the CFs used to produce 35.5 kg of composite is 288€

5.2. LCA of the chemical recycling process in open loop

We conduct the LCA of this new scenario where the carbon fibers are not recovered as long fibers but as shredded fibers. We assume that shredded fibers are assimilated to PAN, the precursor of carbon fibre production and we select this option because the data for PAN is available in the Ecoinvent database, other types of carbon fibers are not available.

Fig. 10 is the network of the recycling processes with recovery of long CFs. The avoided GWP for the recycling process is −108 kg CO$_2$eq. The avoided impacts results associated to the recovered short CFs (−128 kg CO$_2$eq) and the epoxy-thermoplastic (−97 kg CO$_2$eq) will be allocated to another LCA that considers the reuse of these materials. In a previous study it was assumed to recover the CFs maintaining the same properties of the virgin CFs used, but this technology is still under investigation and is not ready yet. In this case, the avoided impact for CFs were accounted in closed loop (Fig. 1) and final impact for the virgin CFs accounted in closed loop was 20,4 kg CO$_2$eq while the initial amount of GWP for virgin CFs is $2.02 \cdot 10^3$ kg CO$_2$eq (see Fig. 10).

In summary, both open loop and closed loop scenarios must be considered, as diagrammed in Fig. 11: the open loop recycling of the CF-epoxy composite to recover the shredded CFs and the epoxy thermoplastic (Fig. 11a); the manufacture of a new CF-thermoplastic composite by compounding and injection moulding the recovered CFs and the epoxy-thermoplastic including the thermal recycling of their products is a closed loop (Fig. 11b).

Experimental processing and testing of this new CF-thermoplastic composite is currently ongoing. Some results are reported in paragraph 6. The full LCA (including product A life cycle and product B life cycle) will be completed once all primary data on manufacturing and recycling of product B will be collected.

6. Experimental lab work on the CF-thermoplastic composite manufacturing

Experimental manufacturing and testing relating this new CF-thermoplastic composite is addressed here by running some compounding experiments on the recovered materials after the chemical recycling process. The recycled materials were obtained by composite panels presented in previous papers that showed a tensile strength and modulus of 518.91 ± 23.02 MPa e 23.71 ± 2.87 GPa (Cicala et al., 2018). However, the possibility to further recycle the products obtained therefrom is not considered here as it will be analyzed in a subsequent paper.

6.1. Materials and samples preparation

The CF-thermoset was processed into a thermoplastic according to the procedure outlined previously (La Rosa et al., 2016). The obtained thermoplastic (reTP) was processed in a twin screw Lab Compounder KETSE 20/40 D (Brabender® GmbH & Co. KG, Duisburg Germany) with the addition of short carbon fibres obtained from recycling. Different carbon fibre content was produced (Table 8). The operating mixing conditions used for compounding are reported in Table 9. The obtained compounds were made into pellets and shaped into dog bone samples by injection moulding, by using a MegaTech H7/18−1 Injection Moulding machine (Tecnica-Duebi, Fabriano, Italy), setting the process parameters reported in Table 10. The dogbone samples were obtained according to dimensions specified by ASTM D638.

The recycled thermoplastic obtained from the recycling of the CF-thermoset was compounded successfully with short recycled carbon fibers. The compound obtained was processed using an injection moulding machine easily up to a CF content of 30 wt%. The samples obtained are shown in Fig. 12.

6.2. Tensile testing

The samples obtained by injection moulding were tensile tested on a universal testing machine Instron 5985 (Instron, Milan, Italy) equipped with a load cell of 10 kN, in strain control at speed of 2 mm/min. The measurements were conducted on dog bone-shape specimens (ASTM D638, Type IV), considering 5 replications for each mixing ratio, for a total of 20 runs. The responses observed were the Yield Stress, the Young's Modulus and the Elongation at Yield. For system control and data collection Blue Hill 3.61 software (Instron, MA, USA) was used.

Differences in mechanical results were statistically analyzed by one-way analysis of variance (ANOVA) using Minitab 17 software (Pennsylvania, USA).

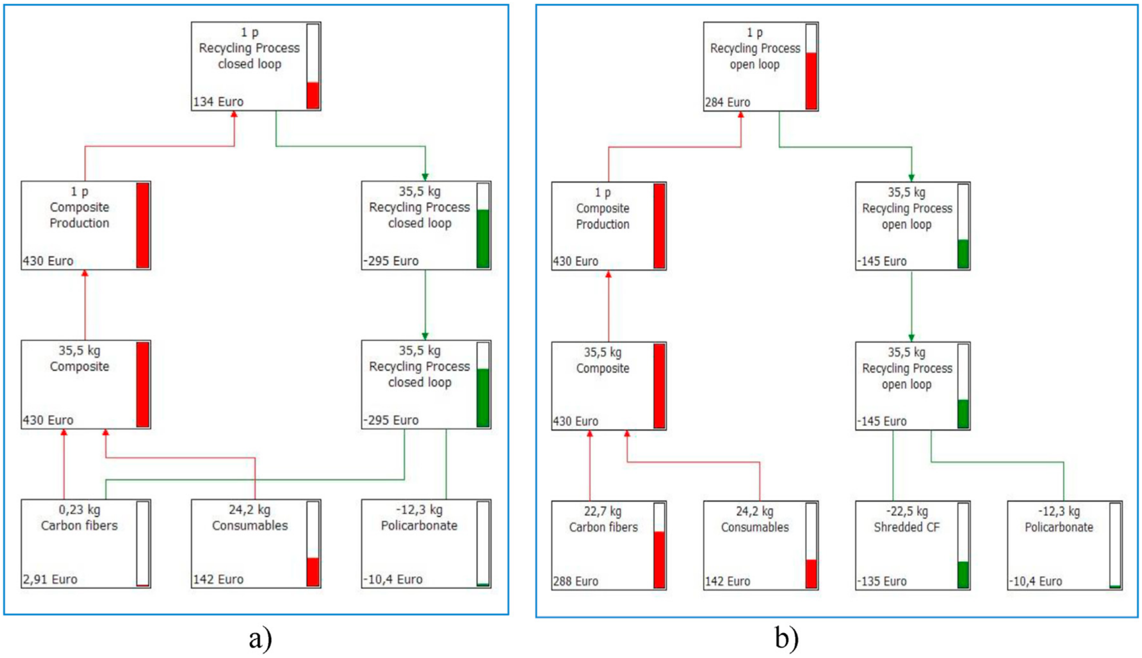

Fig. 9. a) LCC of the closed loop system scenario. The recycled carbon fibres are assumed to maintain the same properties and the same costs of the virgin CFs; b) LCC of the open loop system scenario. The recycled carbon fibers are assumed to have different properties and cost.

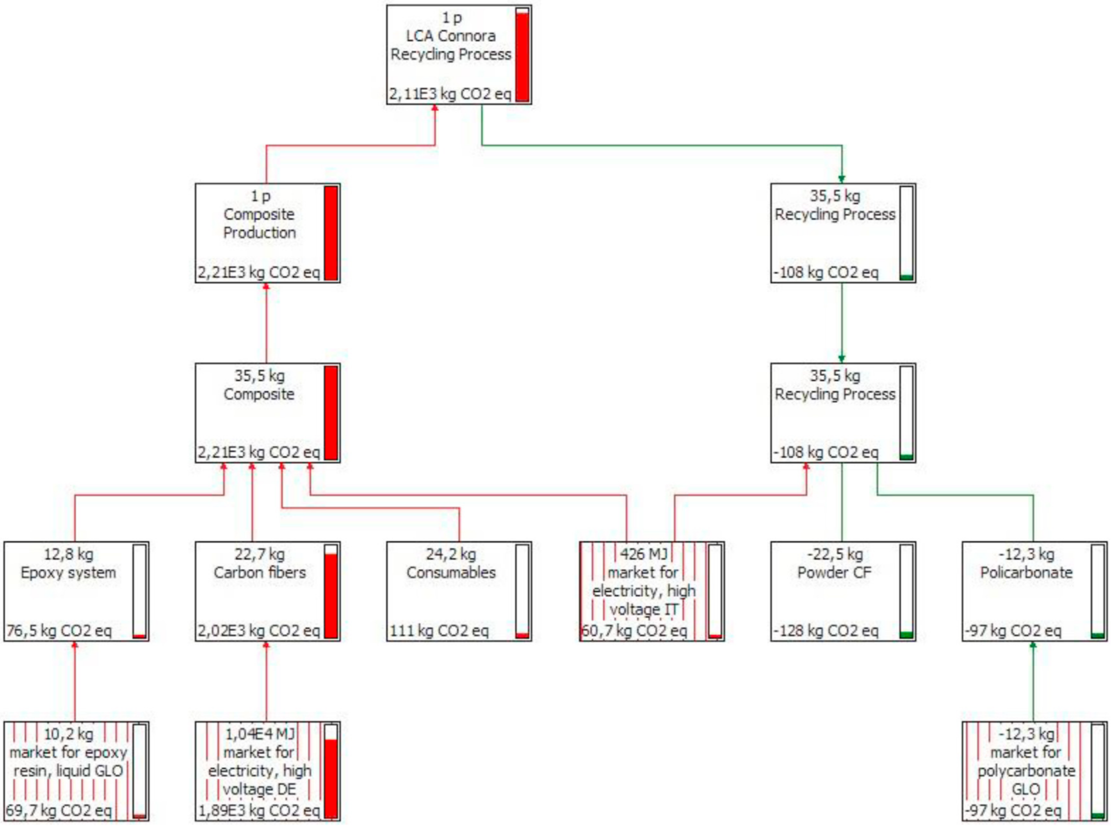

Fig. 10. Network of Recycling Process. Scenario of recovering shredded CFs.

The tensile test data and the average tensile stress vs displacement curves are reported in Table 11 and in the individual plots of Fig. 13, respectively. The Yield Stress varied in the range between 38.45 MPa and 97.52 MPa. The Young's Modulus varied in the range between 2.03 and 13.12 GPa. The Elongation at Yield varied in the range between 3.15 and 1.43%. ANOVA analysis confirmed that the

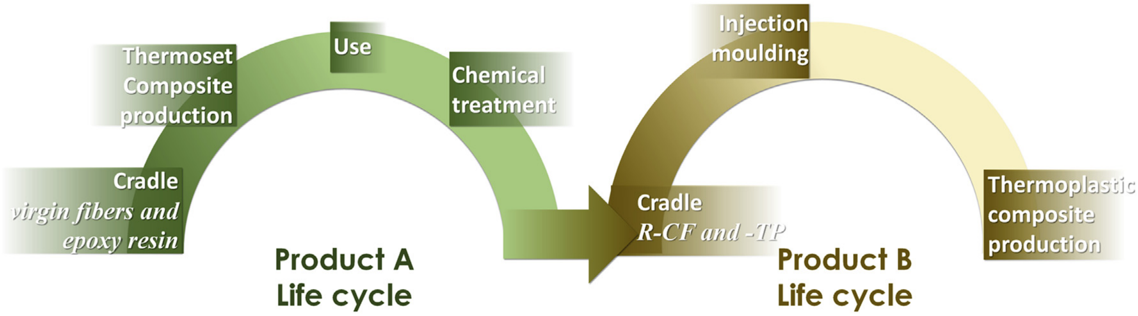

Fig. 11a. Open loop recycling.

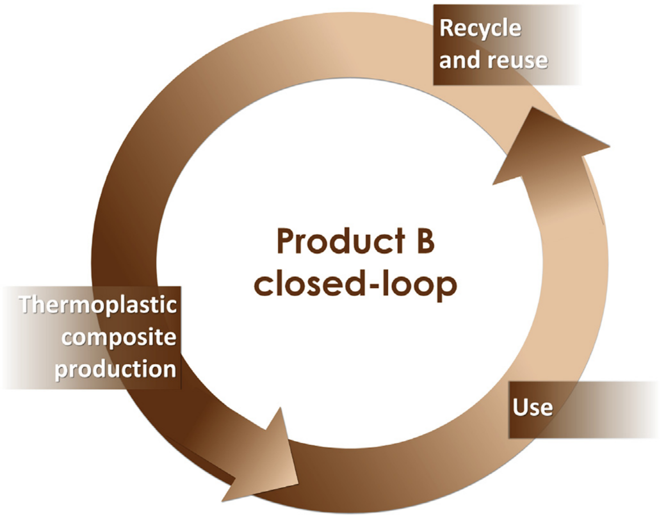

Fig. 11b. Closed loop recycling.

Table 8
Mixing ratios reTP and CFs.

ID Sample	CFs Content [%wt]
reTP	0
reTP-CF10	10
reTP-CF20	20
reTP-CF30	30

Table 9
Extrusion working conditions.

Parameter	Unit	Value
Zone 1 Temperature	[°C]	180
Zone 2 Temperature	[°C]	190
Zone 3 Temperature	[°C]	210
Zone 4 Temperature	[°C]	210
Zone 5 Temperature	[°C]	210
Zone 6 Temperature	[°C]	190
Zone 7 Temperature	[°C]	190
Pressure	[bar]	7
Pump flow rate	[cm^3/min]	14
Extrusion rotation	[rpm]	7

Table 10
Injection moulding working conditions.

Parameter	Unit	Value
Drying temperature	[° C]	80
Drying time	[h]	24
Closing force	[kN]	70
Extractor force	[N]	6800
Injection force	[kN]	27
Cylinder diameter	[mm]	18
Specific pressure	[bar]	1080
Injection speed	[mm/s]	66
Cooling time	[s]	30
Temperature of feed zone	[°C]	20
Temperature of zone 1	[°C]	165
Temperature of Zone 2	[°C]	180
Temperature of cylinder injection	[°C]	180
Temperature of the mold	[°C]	40 °C

observed stress, modulus and elongation means were significantly different for the different contents of CFs (p-value<0.05, Table 4). The trend reported in Fig. 13 shows a very good reinforcing efficiency for the recycled thermoplastic matrix as confirmed by the increasing values with increasing carbon fibre contents for both the yield stress and the modulus. The mechanical properties obtained fall in the range of those reported for virgin CF-thermoplastic compound used in additive manufacturing (Brenken et al., 2018) thus opening a wide range of options as feasible use of compounds obtained (Table 12).

7. Conclusions

In conclusion, previous LCA results that we obtained for the Connora® chemical treatment of recyclable thermoset composite were updated in the present study, as follow:

7.1. Closed loop recycling scenario

The closed loop recycling of CFs generates a reduction of GWP from $2.02\cdot10^3$ kg CO_2eq to 20.4 kg CO_2eq (compared to virgin CFs) and a reduction of costs from 288 to 2.91 € for the chosen functional unit. Unfortunately, the technology to recover CFs with the same properties and the virgin CFs used is still under investigation but it is not ready yet. Therefore, the closed loop recycling scenario is still not realistic for carbon fibers.

7.2. Open loop recycling scenario

The open loop recycling LCA presented in this paper includes the thermoset composite production (starting from virgin CFs and epoxy resin) and the Connora® chemical treatment with the reclamation of short carbon fibers and the epoxy thermoplastic. The avoided impacts accounted in the open loop recycling approach were −128 kg CO_2eq for the short, shredded CFs and −97 kg CO_2eq for the epoxy-thermoplastic. Avoided costs of −135€ for the short, shredded CFs and −10€ for the epoxy-thermoplastic were also accounted, but these avoided impacts

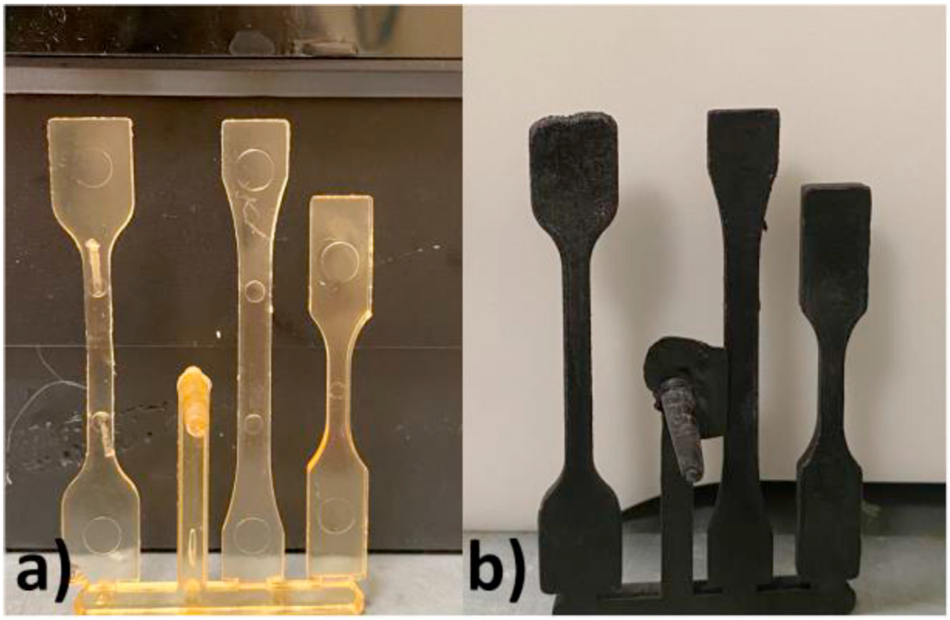

Fig. 12. Dogbone specimens obtained from injection moulding: a) neat recycled thermoplastic (reTP); b) recycled thermoplastic mixed with 30 wt% of recycled CF (reTP-CF30).

Table 11
Tensile test results.

ID Sample	Young's Modulus [GPa]	Yield Stress [MPa]	Elongation at Yield [%]
reTP	2.14 ± 0.13	38.97 ± 1.18	2.99 ± 0.13
reTP-CF10	3.85 ± 0.49	45.02 ± 3.37	2.08 ± 0.17
reTP-CF20	7.48 ± 0.68	69.46 ± 5.82	2.04 ± 0.32
reTP-CF30	12.29 ± 1.03	92.04 ± 4.44	1.67 ± 0.19

Table 12
ANOVA table results.

Response	p-value	R-sq	R-sq(pred)
Young's Modulus	0.000	97.78%	96.29%
Yield Stress	0.000	96.98%	95.13%
Elongation at Yield	0.000	86.62%	78.65%

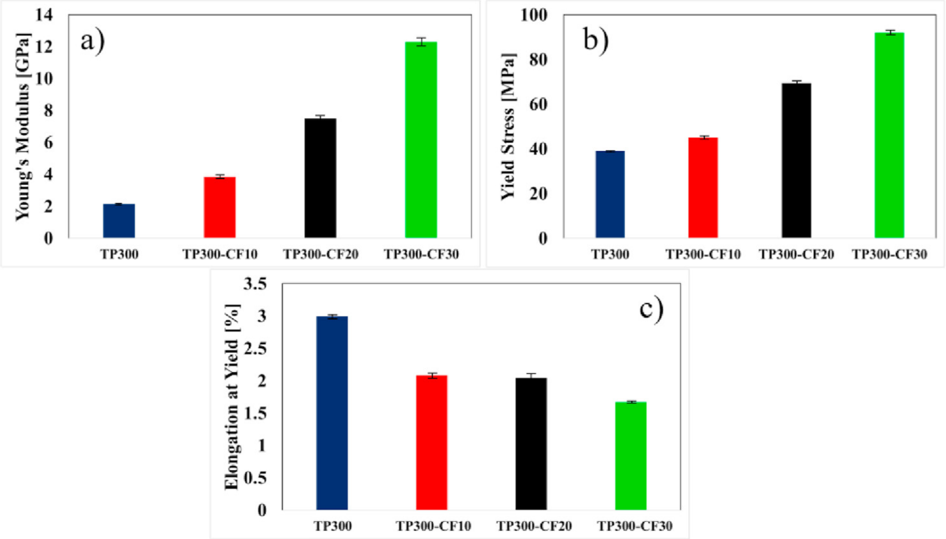

Fig. 13. Individual value plots for: a) Young's Modulus; b) Yield Stress; c) Elongation at Yield.

and costs will be allocated in a further LCA that considers the remanufacture of these materials to produce the thermoplastic composite. Finally, regarding the obtained results, the limitation of using proxy materials should be mentioned.

7.3. Experimental process

To demonstrate the feasibility of this process at laboratory scale, the CF-thermoset composite wastes were shredded and chemically separated into short carbon fibers and epoxy-thermoplastic polymer. These materials were compounded and injection moulded to produce a new CF-thermoplastic composite. The tensile tests carried out on the CF-thermoplastic obtained showed mechanical properties with maximum values for the yield stress and the tensile modulus (i.e. 92.04 MPa and 12.29 GPa) that falls in the range of those usually observed for ABS and Nylon reinforced polymers used in additive manufacturing. This finding supports the conclusion that the recycled materials obtained with the chemical process discussed is not only economically and environmentally sustainable, but it is also technically feasible for practical high value applications. As the mechanical test results are promising, the authors are planning to further investigate this process. Laboratory research is already ongoing on the thermal recycling of the new CF-thermoplastic composite to evaluate the numbers of thermal cycles it can withstand before starting to lose the mechanical properties. A cradle to grave LCA and LCC, including the thermal recycling as end of life, will be presented in a future work.

CRediT authorship contribution statement

Angela Daniela La Rosa: Conceptualization, Writing – original draft, Formal analysis, Software, original draft preparation, writing, LCA and LCC analysis, use of Simapro software. **Sebastiano Greco:** Data curation, Software, Inventory data collection and Simapro software use as part of an internship at NTNU under the tutoring of Prof. A.D. La Rosa. **Claudio Tosto:** Data curation, Experimental lab work, sample preparation and testing. Primary inventory data collection, as PhD student under the supervision of Prof. G. Cicala; graphical abstract. **Gianluca Cicala:** Supervision, Writing – original draft, Experimental lab work, supervision on sample preparation and testing. Writing of the lab work contribution. LCA and LCC of a chemical recycling process for waste CFs-thermoset composites. Open loop and closed loop scenarios.

Declaration of competing interest

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

Acknowledgements

G. Cicala acknowledges project Thalassa (PON “R&I″ 2014–2020, grant n. ARS01_00293).

C. Tosto acknowledges Italian MIUR for the funding of his PhD project entitled “AMA” (CUP E67I18001120006).

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