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Cumulative energy demand analysis in the current manufacturing and end-of-life strategies for a polymeric composite at different fibre-matrix combinations

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ABSTRACT

Fibre Reinforced Polymers (FRPs) are finding more applications in different industrial sectors. From a sustainability point of view, a component made of FRPs reduces energy consumption and CO_2 emissions during its use-phase due to the material's lightweight nature. However, the production of these materials impacts the global energy demand significantly. To mitigate this impact, circular economy strategies are essential. This study focuses on a Cumulative Energy Demand (CED) analysis for different End-of-Life (EoL) strategies of FRPs components. Three EoL routes were evaluated: i.e., combustion, recycling and reforming of continuous fibres reinforced thermoplastics. Different fibres and matrices and three Fibre Volume Fractions (FVF) were taken into account. Specifically, Glass Fibres, Carbon Fibres, Polypropylene, and Polyether ether ketone were examined while FVF of 11%, 23% and 45% were evaluated. A Life Cycle Inventory data was built combining literature review and CES Edupack database. The results provided some guidelines for optimising the product's EoL phase in terms of CED reduction underlining the advantages and high competitiveness of the reforming strategy especially if high-performance matrices and/or fibres are processed. Recycling results to be a valuable EoL alternative if FRPs made by high-performance fibres and high FVF are employed while combustion is the more advisable option if low-performance matrices and fibres are used.

1. Introduction

Continuous fibre-reinforced polymers (CFRPs) are innovative materials that combine high strength and stiffness with low density, making them attractive for a wide range of applications. The continuous fibres, if compared to short fibres, provide the material with high mechanical properties, making them stronger and stiffer (Lee et al., 2019). CFRPs are also highly resistant to fatigue and corrosion and have excellent thermal and electrical properties (Ouyang et al., 2022).

The unique properties of composites have made them popular in industries such as aerospace, automotive, construction and sporting goods (Inagaki, 2000). In aerospace, CFRPs are used in aircraft structures and components to reduce weight and fuel consumption. The reduction in fuel and maintenance costs can help to balance out the higher initial cost (Nayak, 2014). In the automotive sector, numerous studies claim that replacing various vehicle components with lightweight materials, such as composites, reduces CO_2 emissions (Ghassemieh, 2011; Jasinski et al., 2015; White, 2013).

While CFRPs offer many advantages, there are also some drawbacks to be faced. Firstly, CFRPs are more costly to produce (Vijayan et al., 2023). Advancements in production methods and increasing demand have, however, resulted in a decrease in costs, making CFRPs more accessible for a wider range of applications (Qureshi, 2022). Indeed, the production of CFRPs components has increased significantly (Meng et al., 2018). The global CFRPs market size reached US$ 2.55 Billion in 2022 expecting the market to reach US$ 3.62 Billion by 2028, exhibiting a growth rate (CAGR) of 5.90% during 2023–2028 (Market research report, 2023).

A second drawback is related to environmental challenges.The production of these materials uses about one-fifth of global energy demand (Verhoef et al., 2018) and, furthermore, increased volumes of CFRPs in today's applications will result in the creation of waste tomorrow. All this waste will have to be managed. In this context, the Circular

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Nomenclature

CE	Circular economy
CED	Cumulative energy demand
CFRPs	Continuous fibre-reinforced polymers
CFRTPs	Continuous fibre-reinforced thermoplastics
CFs	Carbon fibres
GFs	Glass fibres
CF/GF-RTP	Carbon fibre or glass fibre-reinforced thermoplastics
EE	Embodied energy
EoL	End-of-life
EU	European Union
FRPs	Fibre reinforced polymers
FVF	Fibre volume fraction
LCI	Life cycle inventory
PEEK	Polyether ether ketone
PP	Polypropylene
PP-CF	Polypropylene-carbon fibre composite
PP-GF	Polypropylene-glass fibre composite
PEEK-CF	Polyether-ether ketone-carbon fibre composite
PEEK-GF	Polyether-ether ketone-glass fibre composite
H_{mc}	Embodied energy of the composite
H_{ppc}	Energy consumption of the composite pre-manufacturing process
H_{pc}	Energy consumption of the composite manufacturing process
H_{rc}	Recycling energy
H_c	Energy of the formed composite

Economy (CE) paradigm allows the industrial system to be restorative and regenerative from product idea to design in line with the European Union's (EU) 2050 climate neutrality target as part of the Green Deal (European Commission, 2022).

In this perspective, the use of CFRPs is a challenge for their EoL treatments (Ferrara Snider, 2022). In detail, the separation of matrix and fibre is problematic, due to the covalent bonds created in the polymer's chemical structure. Moreover, fillers and additives, added to provide additional properties, increase this difficulty further (Mathieu et al., 2022; Shekarchi et al., 2020; Szewczak, 2021). The types of fibre and matrix affect the EoL treatments to be used. Looking at used matrices, CFRPs can be made either by thermosetting or by thermoplastic polymers. The advantages of composite materials based on thermoplastics matrix is the process reversibility (Aiswarya et al., 2022; Krivonogov et al., 2019; Snyder et al., 2022; von Freeden et al., 2023). These matrices are reinforced mainly by continuous Glass, Carbon and Other fibres (Correia et al., 2015; Vallée et al., 2013).

The present work focuses on CE approaches of continuous fibre-reinforced thermoplastics (CFRTPs). CFRTPs estimated at US$948.9 Million in the year 2022, is projected to reach a size of US$1.5 Billion by 2030, growing at a CAGR of 6% over the analysis period 2022–2030 (Research and Markets, 2023).

Specifically, exploiting the hot re-formability of thermoplastics, three different EoL routes were analysed: combustion, recycling and reforming. These strategies were investigated for different matrices and fibres combinations. The analysis was performed using a methodology available in literature (Suzuki and Takahashi, 2005; X. J. Zhang et al., 2020) quantifying the primary energy demand, i.e., the Cumulative Energy Demand (CED), of the different CFRTPs from cradle-to-grave. In detail, the impacts of two reinforcing materials such as carbon fibres (CFs) and glass fibres (GFs) and two types of thermoplastic matrices such as polypropylene (PP) and polyether ether ketone (PEEK) were evaluated. The materials were combined to obtain four different composite configurations. Specifically, CFs and GFs were analysed because they are the more and the less valuable reinforcements, respectively. In addition, CFs are worth considering the high cost of the raw material, the technology involved in their development and the expected highest growth in the next few years (Stratview research, 2023). These aspects deserve attention in a closed-loop economy perspective (Stieven Montagna et al., 2022). GFs, instead, allow manufacturing the most common composites that constitute more than 95% of production mainly for the transport industry (automobile, railway) and for the electrical construction (Hsissou et al., 2021). Simultaneously, PEEK and PP were chosen as performing and poor matrices, respectively. In particular, PEEK exhibits a distinctive combination of mechanical and electrical properties at elevated temperatures allowing promising applications in different fields. Furthermore, these applications can further evolve by incorporating functional fillers and fibres (Wiley, 2022). On the other side, polyolefins, whose PP belongs to, are widely used for fabricating reinforced composites (Gogoi et al., 2022). Finally, three fibre volume ratios were analysed.

The variables were selected considering the differences in their impact in terms of Embodied Energy (EE). The EE of raw materials refers to the energy required to extract, process and transport the raw materials used in the manufacturing of a product or system. It represents the energy, measured in MJ/kg, consumed from the initial extraction of the resources from the earth to their arrival at the manufacturing site (Ashby, 2020). Furthermore, a detailed inventory analysis was carried out, as, at present, the study of the energy impacts of the composite materials is still too limited and uncertain (Miller, 2021).

The objective of this study is, therefore, twofold. On the one hand, it aims to provide a comprehensive data record useful for composite partitioners, and on the other hand, more importantly, to present a CED analysis of the three highlighted different EoL routes. These two objectives can be considered interconnected, as the dataset obtained in the former was used to achieve the latter. Owing to that, section 2 provides an overview on the production and recycling processes of CFRTPs pointing out the ranges for each energy aliquot. To be more specific, the literature review was developed in order to provide the energy demand of each step of CFRTPs component life cycle. Section 3, instead, describes the definition of goal and scope, functional unit, boundary conditions and methodology used for the case study analysis, whose results and future developments were discussed in sections 4 and 5 according to the standard ISO 14040 (Klüppel, 2005).

2. Investigated manufacturing and EoL strategies

CFRTPs manufacturing involves different technologies and process routes. In the field of thermoplastic composites, the typical manufacturing processes could be autoclave, compression moulding, cold press moulding, automated tape laying, while in the case of thermosets there are spay-up, pressure bagging, microwave curing, vacuum-assisted resin transfer moulding, as stated by (Lunetto et al., 2023). In this work, the autoclave manufacturing process was considered. The use of an autoclave is a common method in the manufacturing process of composite thermoplastics (Ageorges et al., 2001). The autoclave process for composite thermoplastics offers advantages such as uniform compaction, improved consolidation and enhanced fibre impregnation (Fernández et al., 2003). The specific autoclave parameters and process conditions may vary depending on the composite material, part design, and manufacturing requirements (Clancy et al., 2019). Regarding the impact of autoclave manufacturing, looking at the quantification of the energy, lack of information was evidenced by several articles (Forcellese et al., 2020; Lunetto et al., 2023; Stoiber et al., 2021).

As regards the EoL of CFRTPs, landfill is still the most widely used disposal method worldwide (Krauklis et al., 2021). Anyway, the EU's waste Directive introduced restrictions on landfilling of all waste suitable for recycling from 2030 (European Commission, 2021). These results are needed to develop more sustainable routes for a CE of CFRTPs, which can be disposed of, recycled or reformed, through different methods. Commonly, composite wastes are disposed via combustion, such as an incinerator, generating ash and creating, in any case, an environmental impact. This ash can only be landfilled as inert waste, which is detrimental to the CE progress. Another disadvantage is that when heat is converted into electricity, an efficiency of only 35% can be achieved. However, burning coal in the furnace is a much better option than burning CFRP (Krauklis et al., 2021).

Recycling of CFRTPs can be performed without separating the fibres from the matrix. Indeed, mechanical recycling processes are based on shredding composites resulting in a negative effect on mechanical properties of the fibres with the main part of CFRTPs' value being lost because of length reduction and a loss of fibre architecture (ELG Carbon Fibre Ltd., 2017; Morici and Dintcheva, 2022). In this case, the recycled fibres are filamentous and unorganised (Pickering et al., 2016). This EoL route, often proposed owing to its low-cost technologies (Kiss et al., 2020; Pegoretti, 2021), results in components characterised by reduced mechanical properties if a direct impregnation of these fibres is executed (Pickering et al., 2016).

If the fibres are not broken during the recycling phases, the CFs can, instead, maintain their tensile strength, with only a few percentage points less than virgin CFs (Lee et al., 2011). Several studies claim that the reduction in CFs mechanical properties depends on the type of carbon fibre and on the recycling process parameters (Oliveux et al., 2015; Jiang and Pickering, 2016). Furthermore, woven recycled CFs exhibit a similar tensile modulus in the principal directions than virgin woven CFs (Pimenta and Pinho, 2012). On the other hand, tensile strength and failure strain of recycled GFs decrease up to 70% in comparison with virgin GFs (Kao et al., 2012).

Chemical or thermal recycling processes allow the fibres to be separated by the matrix preserving the fibre length. Specifically, the chemical process, the so called solvolysis, allows the polymer matrix to be degraded by a solution of acids, bases, and solvents, whose composition must be fine-tuned to the matrix (Jody et al., 2004; Pimenta and Pinho, 2011). After the process, the recycled fibres are cleaned to remove decomposed polymeric composites and solvent residues and reoriented. On the other side, the thermal process, the so-called pyrolysis, decomposes thermally the polymer removing the pyrolytic char on the carbon fibres by an oxidation process permitting the reinforcing materials to be recovered and reused (Naqvi et al., 2018; Krauklis et al., 2021). Several studies have shown that the decomposition process is performed at a temperature range from 350 °C to 700 °C (Abdou et al., 2016; Giorgini et al., 2015; Meyer et al., 2009; Witik et al., 2013), claim that the pyrolysis process has emerged as more efficient and reliable than solvolysis in terms of energy and material recovery.

Recapitulating, before performing one of the recycling processes, above detailed, different preprocessing solutions have to be evaluated obtaining different levels of retaining of the initial fibres architecture and, consequently, of recovering fibre values (Meng et al., 2018; Khalil, 2018; Pillain et al., 2019). Anyway, even after the recycling step, several post-processing stages can be explored to improve the quality of the recycled fibres. For example, wet paper-making (Wong et al., 2010) or realignment techniques, such as HiPerDif process (Pozegic et al., 2020) or different spinning variants (Akonda et al., 2012; Hengstermann et al., 2016; Hasan et al., 2018; Colombo et al., 2023) have been proposed.

Pre-processing, processing and post-processing stages, therefore, have to be considered together to judge the most promising recycling route of CFs or GFs-reinforced polymers. Currently, at least for the authors' knowledge, few studies have taken into account the whole stages (Meng et al., 2017; He et al., 2020). In particular, He et al. (2020), following the LCA methodology, assessed five different recycling routes taking into account pre-processing, processing and post-processing stages proving that a less impactful solution in terms of energy demand is the route, where the woven fibre architecture is not shredded during the pre-processing stage. More in detail, the less demanding energy route was the one in which the retained architecture was obtained by pyrolysis, subsequently, impregnated with resin, without requiring a post-processing stage and directly reusing the woven in production of a new product. This recycling route, first proposed by Pimenta and Pinho (2012), was, therefore, considered in the research herein presented.

Finally, a perfect CE can be achieved, if the polymer matrix is thermoplastic, by reforming the product providing a new life cycle to it. In this context, owing to the thermoplastic matrix fusibility, CFs or GFs-reinforced thermoplastics (GF/CF-RTP) are considered reformable Kiss et al. (2020); Kiss et al., (2020) in their study, revealed that the reverse forming, so-called reforming, is a viable route for CFRTPs. They showed that this method can be also applied to correcting any forming mistake or to reform the product at the EoL. Von Freeden et al. (2023), focused the attention on the reforming process and its effect on composite sheets. They stated that lifespan of CFRTPs could be extended up to 5 processing cycles compared to alternative materials.

2.1. Embodied & manufacturing energy

The analysis of the available scientific literature has shown a great variability in EE values of the raw materials. In Table 1, the embodied energies of the reinforcements and the thermoplastic matrices, analysed in the proposed study, are summarised. The values marked with a star symbol (*) indicate that the level of energy form was not specified in the literature data. Therefore, data reliability could be threatened. Anyway, the risk in the data consistency was already taken into account referring to embodied energies of materials that have a brief scientific history and that are characterised by high variability as also stated by (Ashby, 2020). To mitigate this weakness, the study was performed considering the whole ranges of the detected values.

The fibres need to be produced by specific sub-processes to be combined in yarns, which can be considered as the base unit for the woven construction. In Table 2, these sub-processes are detailed for both CFs and GFs yarns summarising the energy consumption for each phase. Specifically, CFs are obtained by the polymerization of Polyacrylonitrile (PAN) while GFs are produced starting by a molten SiO_2 slurry. In Fig. 1 (a), these different sub-processes are illustrated. According to that, it has

Table 1
Embodied energy of the investigated raw materials.

	Material	Embodied energy (MJ/kg)	Reference(s)
Polymer matrix	PP	72.00–112.00	Song et al. (2009)
		24.00	Granta Design Limited (2023)
		66.00–80.00	Granta Design Limited (2023)
		11.00–27.00	Lunetto et al. (2023)
	PEEK	286.00–315.00	Granta Design Limited (2023)
Reinforcement	CF	183.00–286.00 *	Oliveux et al. (2015)
		272.00–300.00	Granta Design Limited (2023)
		280.00 *	Oliveux et al. (2015)
		1000.00	(Duflou et al., 2009; Witik et al., 2012)
		704.00	Witik et al. (2012)
		1468.00 *	Katsiropoulos et al. (2019)
		286.00–478.00	Lunetto et al. (2023)
		190.00–870.00 *	Liddell et al. (2017)
		521.00–1563.00 *	Oliveux et al. (2015)
		855.00	Witik et al. (2012)
	GF	49.00–54.00	(Granta Design Limited, 2023; Lunetto et al., 2023)
		13.00–32.00 *	Oliveux et al. (2015)
		7.00–16.00 *	Liddell et al. (2017)

Table 2
Sub-processes used to weave the yarns.

Fibres	Sub-processes	Energy consumption (MJ/kg)	Reference(s)
CF	1.PAN Polymerising	0.00–156.00	Song et al. (2009)
	2.PAN spinning	2.60	Liddell et al. (2017)
	3.Oxidation	142.00–427.00	
	4.Finishing	35.00–75.00	
GF	1.Molten slurry of SiO_2	1.30–2.50	
	2.Melting	3.40–9.10	
	3.Yarn Spinning	2.60	
	4.Finishing	0.90–1.90	

to be clarified that the energy consumption is a value that is obtained by converting the wasted energy required to execute a specific process, and quantified by the absorbed electric energy, measured in MJ, into MJ oil equivalent, which depends on the employed country's energy mix. For the performed analysis, the energy consumption refers to the European average energy mix (Ashby, 2020).

Once the yarns are obtained, these have to be weaved to achieve the fabrics. Therefore, the yarns are wrapped and oriented by specific crimp angles to create the desired woven fabric. This can be performed using techniques such as weaving, knitting, or braiding methods. The energy used in this manufacturing phase was estimated by (Song et al., 2009). Specifically, this energy consumption was quantified at 2.90 MJ/kg. The woven fabric, subsequently, has to be impregnated with the thermoplastic resin to obtain the prepregs by applying heat and pressure to soften the matrix, allowing it to impregnate and bond with the reinforcement. This can be executed using techniques like hot pressing, hot melt infusion, or thermoforming (Fig. 1(b)). For the impregnation, the energy required for the PP and the PEEK matrices was quantified in the range of 20.80–23.00 MJ/kg and 25.30–27.90 MJ/kg, respectively (Song et al., 2009). The prepregs must be finally manufactured by the autoclave moulding process. Also, for this working step, the energy consumption values vary depending on the characteristics of the polymer matrix and its melting temperature. Furthermore, the production volume must be considered in the assessment of energy consumption (Suzuki and Takahashi, 2005). Considering the investigated thermoplastics, the process energy values were found in literature and listed in Table 3.

Finally, the cutting phase necessary to finish the demoulded parts obtained after autoclaving requires energy ranging from 0.10 to 1.40 MJ/kg as detected in the literature (Bianchi et al., 2021) (Fig. 1(c)).

2.2. EoL considerations

As already mentioned, the increasing use of composite materials in several sectors results in problems of waste management (Altay et al., 2018; Karsli and Aytac, 2013). When a material is recycled, it often requires less energy than extracting and processing the raw material from scratch. For this reason, different recycling methods have been proposed (Giorgini et al., 2015, 2020; Oliveux et al., 2015; Jiang et al., 2015; Khurshid et al., 2020; X. Zhang et al., 2020; Tapper et al., 2020; van de Werken et al., 2020; Qureshi, 2022). In this work, combustion, recycling (thermal/chemical) and reforming were analysed. Specifically, about the recycling process, (Table 4), the solvolysis (chemical recycling) consists in the removal of the thermoplastic matrix by dissolution in a proper solvent. The advantages of chemical recycling over thermal recycling, is that lower temperatures are generally

Table 3
Energy consumption required for the autoclave moulding process.

Thermoplastic matrix	Melting temperature (°C)	Energy consumption (MJ/kg)	Reference(s)
PP	160	141.00	Katsiropoulos et al. (2019)
		111.36–141.00	Vita et al. (2019)
PEEK	340	163.68	Katsiropoulos et al. (2019)

Table 4
Energy consumption of the investigated EoL routes.

EoL phase	Method	Material	Energy consumption (MJ/kg)	Reference(s)
Combustion	Incinerator	PP/PEEK	30.50–32.00	Tapper et al. (2020)
			32.00–33.60	Granta Design Limited (2023)
Recycling	Pyrolysis	PP	2.80–30.00	Tapper et al. (2020)
		PEEK	23.98–63.00	Katsiropoulos et al. (2019)
	Solvolysis	PP	15.00–64.00	Tapper et al. (2020)
		PEEK	61.00–93.00	
Reforming	Thermo-forming	PP	3.23	Lee (2021)
		PEEK	3.82	
		PP	28.68	
		PEEK	45.29	

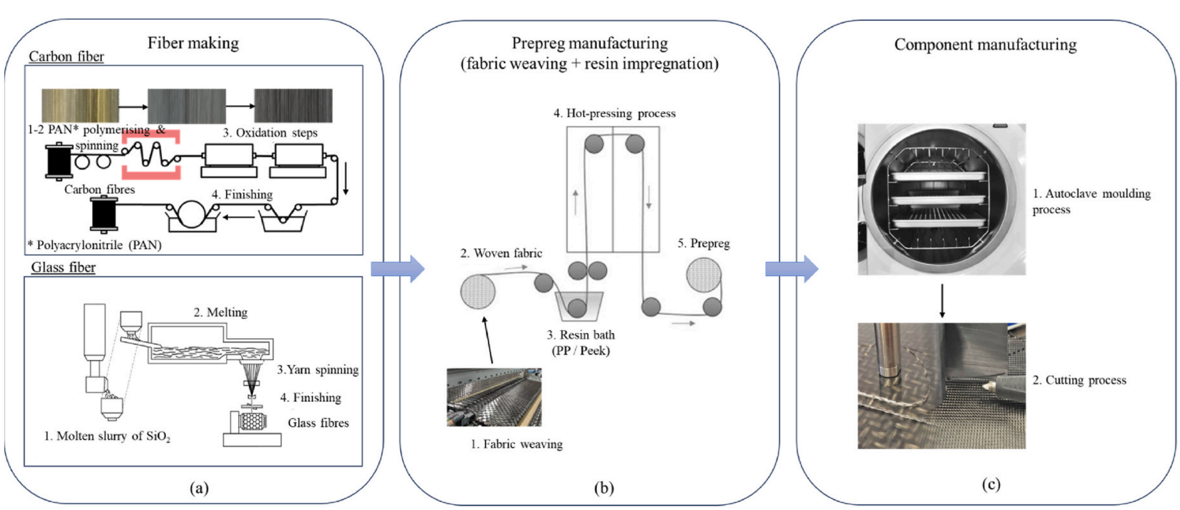

Fig. 1. The whole production phases of the target component made of CFRTPs: (a) Yarn, (b) prepreg and (c) component manufacturing.

required to degrade the polymeric matrices (Oliveux et al., 2015) reducing possible damages on the recovered fibres allowing recovery of both the polymer matrix and the full-length fibres (Cousins et al., 2019). Anyway, a limited number of studies is still available in the scientific literature on the chemical recycling of CFRTPs (Pegoretti, 2021), which can be performed by using a wide spectrum of solvents and catalysts that significantly affect the environmental impact of the EoL phase. For this variability and data solidity, the solvolysis was not further explored leaving its analysis to a following research step.

In detail, the process energy of three different EoL phases was outlined in Table 4. Combustion, where both matrix and fibres are wasted, recycling by pyrolysis conserving the full architecture of the woven fabric, and reforming, where the whole materials are saved for a new manufacturing phase, were considered in the executed study. The impact of solvolysis was reported just for further data information deserving, as above highlighted, a specific in-depth analysis looking not just at this EoL's energy consumption, but also at the impacts of employed solvents and catalysts.

The values used in Table 4 were extracted from literature and Cambridge Engineering Selector Edupack database (CES) (Granta Design Limited, 2023). In addition, a cleaning step and a reorientation phase are required in the recycling phase. For what concerns the cleaning step, the energy consumption is 8.73 MJ/kg (Kooduvalli et al., 2022). The cleaning phase is necessary to purify the reinforcement (GF/CF) to make it ready for a new life cycle. In the reforming route, instead, the reforming process consists of a thermal forming phase that consists of heating the component and a consolidation phase known as calendaring. Considering the polymer matrix of PP (melting temperature 160 °C) and PEEK (melting temperature 340 °C), a heat-assisted forming tool step is required during thermal re-forming. The energy consumption for these reforming phases is 6.882 MJ/kg for PP and 15.122 MJ/kg for PEEK (Lee,2021). Finally, a heat treatment is required during the calendering process. The energy consumption for this process step is 3.354 MJ/kg and 6.215 MJ/kg if the composite is made, respectively of PP or PEEK (Lee, 2021). The different EoL routes are described in Fig. 2.

3. Material and methods

Four different material combinations were analysed using GFs and CFs reinforcement fabrics, 2/2 twill balanced weave (Doris, 1989), and PP and PEEK polymeric matrices. A low performing polymer matrix (PP) and one with high-performance (PEEK) were taken into account. The same consideration was made for the fibres' selection, being GF and CF known as a low and a high-performance reinforcement, respectively. Furthermore, different percentages of reinforcement were analysed. Specifically, the investigated Fibre Volume Percentages (FVF) are: 45% (FVF 1), 23% (FVF 2) and 11% (FVF 3). These reinforcement percentages were considered because 45% is close to the upper limit of reinforcement that can be achieved in a composite while 11% is close to the lower limit (below which reinforcement fails to improve the performance of the composite (Mallick, 2007). Finally, 23%, besides being a typical value for reinforcements within composites, was chosen as this is a volume percentage that is almost double of 11% and half of 45%. The CED impact of the various composite sheets was evaluated considering three different EoL routes. The research was performed without considering the changing of performance between virgin and recycled or remanufactured materials. According to Von Freeden et al. 2023, longer and multiple use of the composite material in high quality condition was proved. However, the demonstration was performed at laboratory level and needs further studies to be evaluated considering additional phenomena (Mercier et al., 2008). Hence, a CED analysis of the materials

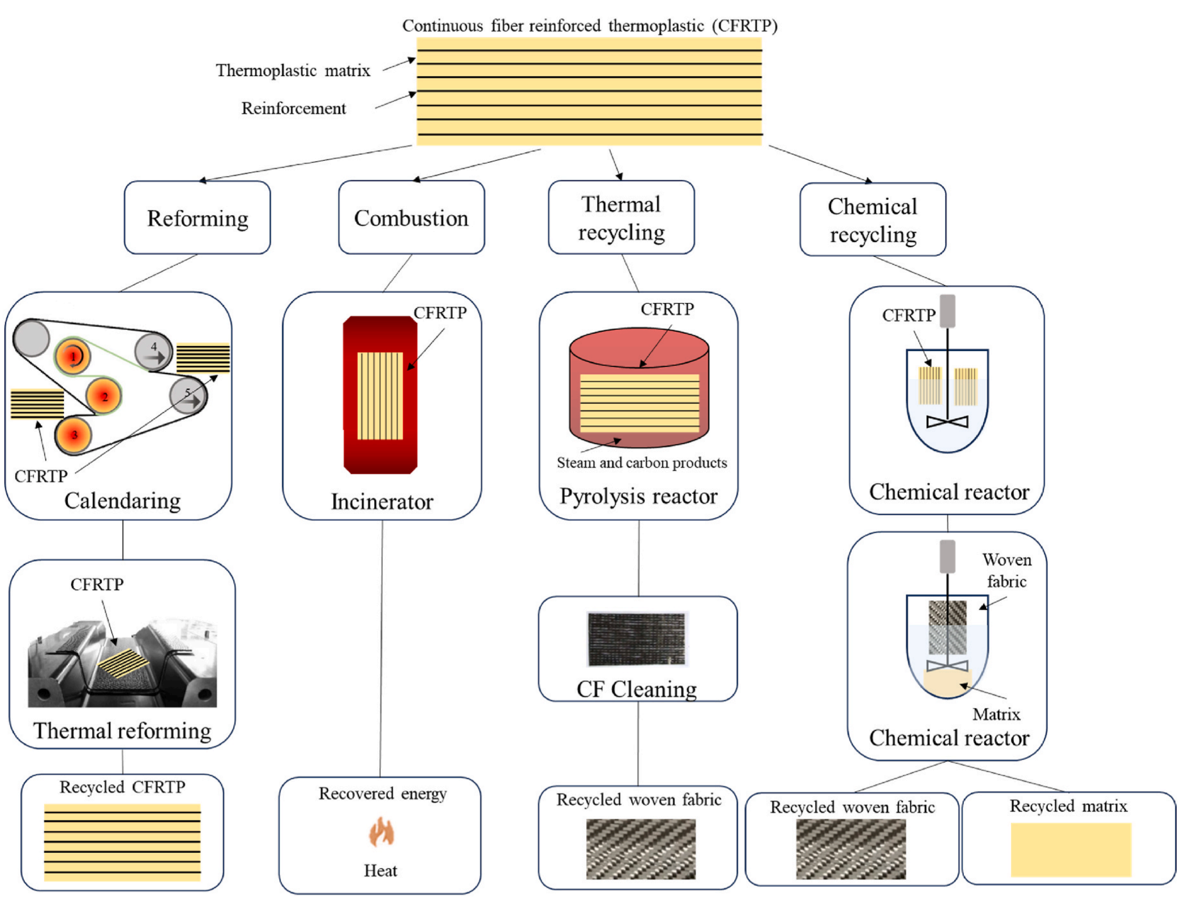

Fig. 2. Main steps to move from the initial composite/scrap to the final recycled product for reforming, combustion, and thermal and chemical recycling.

and processes was carried out, according to the type of reinforcement, matrix, and their percentage in the composite materials. The product system is a component made of CFRTPs, the so-called target component, which is characterised by a volume of 78.4 mm^3. Specifically, composite sheet blanks with dimensions of 280mmx280mmx1mm were considered. The densities of the matrices and of the reinforcements are summarised in Table 5.

3.1. Goal and scope definition

The aim of the study is to characterise the environmental impact, from a CED point of view, of one target component manufactured by each EoL route. The results can be used to compare the CED of EoL routes analysed in the study. The aim is to analyse these impacts for each route to provide guidance for the selection of the most energetically friendly EoL strategy with varying production scenarios. The functional unit chosen for this study is the EoL processing of one unit of the target component (composite sheet blank with dimension 280mmx280mmx1mm). Indeed, the general idea is to compare the EoL processing, looking at the primary energy used from cradle to grave for each of the highlighted EoL scenarios. The benchmark EoL scenario was the so-called conventional open loop process allowing a CED assessment of a component that is processed by combustion. The second scenario is a partially closed loop because the woven fabric, thermally recycled, once cleaned and reoriented, needs, subsequently, to be impregnated with the virgin thermoplastic resin to obtain the prepregs. The third scenario envisages a whole closed loop involving the recovery of the entire component, with the possibility of changing its original shape for a new use. The three detailed manufacturing scenarios are summarised in Fig. 3.

In the study, a recycled content approach was applied. This approach, also called the 100:0 or the recycled content, considers that the environmental impacts of the production phase for a product are attributed to the first use of this product and follows the “polluter pays” principle (Gervasio et al., 2018). The second use of the product only bears the environmental impact of collection and the preparation of the product for its subsequent use. In some cases, the collection is also attributed to the first use of the product. However, the materials used for the second time bear no environmental burden from the primary production process (Frischknecht, 2010; Obrecht et al., 2021).

3.2. System boundary and main assumptions

Concerning the metric for the comparison of the environmental impact, CED (MJ) was used. Indeed, since the first LCA studies, CED has been one of the considered key indicators (Frischknecht et al., 2015). The adopted system boundary includes raw material extraction, product manufacture and EOL, as schematised in Fig. 4. The analysis does not include the use phase’s contributions. Indeed, the use phase was neglected, being common to the three processes examined. Furthermore, the impact of transport between process units was not taken into account, as it is assumed to be the same between process units and between the analysed scenarios. In the following paragraph, some details about assumptions made to deal with electrical energy demand and material

Table 5
Density of the investigated materials.

	Polymer matrix		Reinforcement	
	PP	PEEK	CF	GF
Material density (kg/m^3)	912.50	1320.00	1900.00	1857.00
Reference(s)	Su et al. (2019)	Na et al. (2018)	(Sezgin et al., 2017)	Poso et al. (2021)

The target components’ configurations analysed, namely PP-CF, PEEK-CF, PP-GF, PEEK-GF are summarised in Table 6.

Table 6
Component features expressed in kg at the three investigated FVF.

PP-CF	FVF 1	FVF 2	FVF 3
Fibre mass	0.067	0.034	0.017
Matrix mass	0.039	0.055	0.063
Component weight	0.106	0.089	0.080
PEEK-CF			
Fibre mass	0.067	0.034	0.017
Matrix mass	0.057	0.080	0.092
Component weight	0.124	0.114	0.108
PP-GF			
Fibre mass	0.065	0.032	0.016
Matrix mass	0.039	0.055	0.063
Component weight	0.104	0.088	0.079
PEEK-GF			
Fibre mass	0.065	0.032	0.016
Matrix mass	0.057	0.080	0.092
Component weight	0.122	0.113	0.108

scraps, if present, were specified. Regarding the process’s electric energy demand, it was converted into primary energy source consumption by considering an average efficiency of 36% to account for the energy generation and the transmission losses (Ashby, 2020).

As far as material waste is concerned, all the material creates an environmental impact for the conventional open loop process because the whole component at the end of its life is processed via incinerator. Whereas for the partially closed loop approach, all the fibres were considered without any degradation due to the thermal recycling phase while the matrices become waste and a new quantity of thermoplastic, PP or PEEK, must be added in the process. Finally, for the whole closed loop, on the other hand, there was no material’s environmental impact, as all material is reused and reformed. Furthermore, in this case the mechanical performances of the material remain unchanged (von Freeden et al., 2023).

3.3. Life cycle inventory

LCI data were generated using different approaches. Data from scientific literature and CES Edupack database (Granta Design Limited, 2023) were used. A great variability in the parameters of EE was found in the scientific data, as previously shown in Tables 1–2 Therefore, the analysis was conducted considering the lower, the average and the higher values as described in Table 7, where the various energy contributions were organised. In detail, H_{mc} gathers the EE values necessary to obtain both matrices and fibres and the sub-processes energy required to combine fibres in yarns. H_{ppc} collects the energy for weaving the yarns, achieving the fabrics and the energy for their impregnation to get the prepregs. H_{pc} lists the energy required to form the prepregs, i.e., the manufacturing energy, including the cutting phase. According to this aliquot, the autoclave manufacturing process assumes two energy values depending on the different process temperatures of PP and PEEK (Table 3). Indeed, the process temperature is closely related to the process energy (Katsiropoulos et al., 2019; Vita et al., 2019). Finally, H_{rc} reports the EoL energy taking into account all the routes analysed. As already written, the data related to solvolysis were reported to provide a complete inventory data, even if pyrolysis was the only considered in the study, being the most energy efficient recycling process from a CED’s point of view (Kawajiri and Kobayashi, 2022) and considering the lack of data for chemical recycling of CFRTPs. Furthermore, the processes of reforming and reconsolidation were, instead, obtained by the energy absorption of the employed machines considering the composite’s reprocessing temperature (von Freeden et al., 2023).

3.4. Life cycle energy demand quantification

The methodology proposed by Suzuki and Takahashi (2005) to quantify the CED was applied to perform a comparative analysis of the

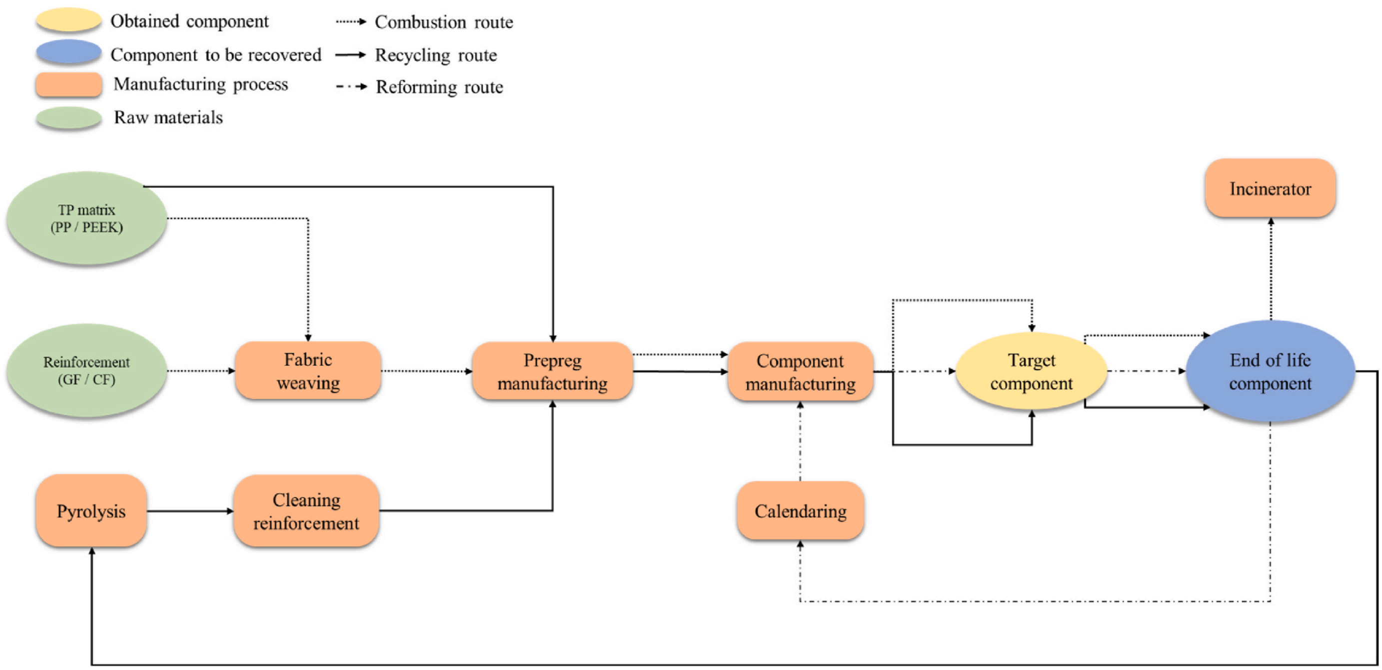

Fig. 3. Detail of the analysed EoL routes.

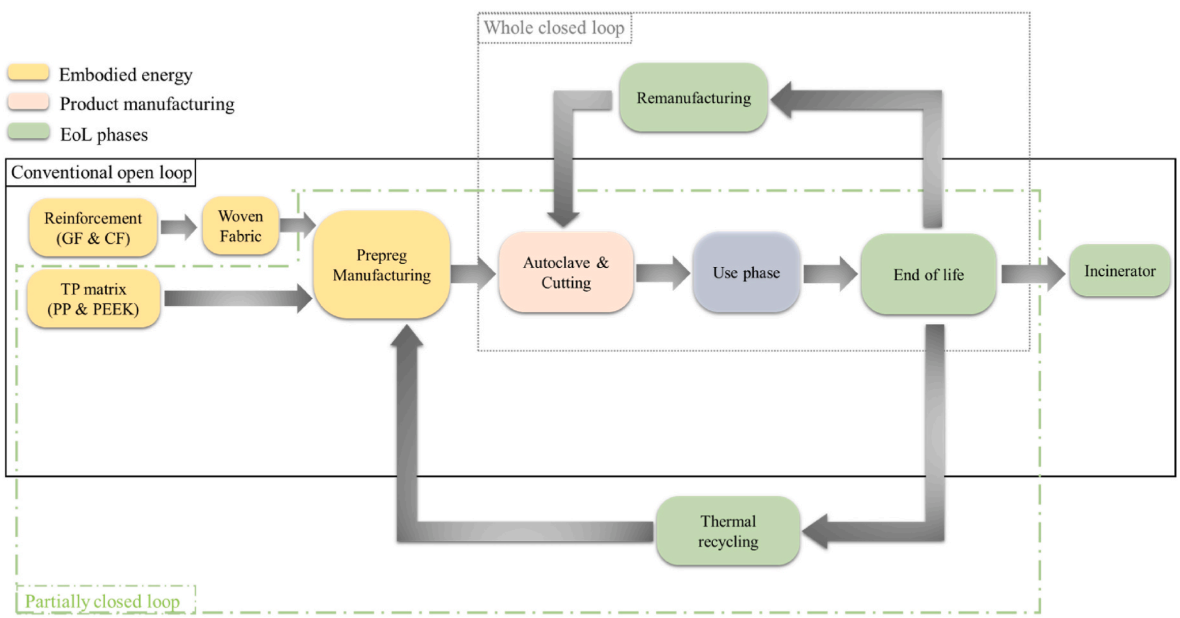

Fig. 4. The adopted system boundaries.

case study's environmental impact, described in Section 3. The life cycle primary energy demand quantification analysis was performed using Eqs. (1)–(5). The CED analysis of the target component assesses the environmental impact during its life cycle. The method considers the EE of the composite material (H_{mc}) based on the weight of the polymer matrix fraction and of the reinforcement as summarised in Eq. (1). The EE of the component is calculated as the mass fraction of the matrix (mf_m) and of the fibres (mf_f) multiplied by the EE of the matrix (H_{mm}) and of the fibres (H_{mf}), respectively. The contributions of the processes used in pre-manufacturing and manufacturing are included through the energy consumption parameters (H_{pp} and H_p) (Eqs. (2) and (3)). The overall energy of the formed composite product is quantified by H_c (Eq. (4)).

$$H_{mc} = mf_m \bullet H_{mm} + mf_f \bullet H_{mf} \tag{1}$$

$$H_{ppc} = mf_m \bullet H_{pp1} + m_c \bullet H_{pp2} \tag{2}$$

$$H_{pc} = m_c \bullet \left(H_{p1} + H_{p2} + \ldots + H_{pn}\right) \tag{3}$$

$$H_c = H_{mc} + H_{ppc} + H_{pc} \tag{4}$$

Eq. (4) can be used to quantify the CED of the target component for the combustion route (conventional open loop) adding just the contribution of H_{rc}. Being the developed analysis based on the recycling content approach, Eq. (5) was instead used to calculate the component's CED in the recycling and reforming routes (respectively, partially and

Table 7
Life cycle inventory data.

	Processes	Energy consumption (MJ/kg)		
		Low	Ave	High
H_{mc}	CF embodied energy	608.78	722.39	836.00
	GF embodied energy	23.00	28.50	34.00
	PP embodied energy	43.25	52.00	60.75
	PEEK embodied energy	283.01	290.26	297.51
	CF manufacturing	47.75	132.63	217.50
	GF manufacturing	1.75	2.90	4.05
H_{ppc}	PP manufacturing	20.80	21.90	23.00
	PEEK manufacturing	25.30	26.60	27.90
	Fabric manufacturing	2.54	2.60	2.67
H_{pc}	Autoclave (PP)	55.68	90.93	126.18
	Autoclave (PEEK)	65.60	114.64	163.68
	Cutting phase	0.05	1.76	3.47
H_{rc}	Incinerator	30.50	31.25	32.00
	Pyrolysis (PP)	3.00	16.50	30.00
	Pyrolysis (PEEK)	23.98	43.50	63.00
	Cleaning	0.00	1.76	8.73
	Re-forming (PP)	10.11	10.11	10.11
	Re-consolidation (PP)	32.03	32.03	32.03
	Re-forming (PEEK)	18.94	18.94	18.94
	Re-consolidation (PEEK)	51.51	51.51	51.51

whole closed loops):

$$H_{Recycling\ content\ approach} = R \bullet \left[\left(H_c - \left(H_{rc} \bullet mf_m + H_{rc} \bullet mf_f \right) \right] \right. \quad (5)$$

where, R is the fraction of the recycled material. R value is equal to 100% for the reforming route while, in the recycling strategy just the fibres are completely recycled.

4. Discussion of results

The CED values for the four material configurations, listed as: PP-CF, PEEK-CF, PP-GF, PEEK-GF, for each FVF and EoL strategy are reported in Fig. 5.

The impact of the different EoL strategies on CED depending on the type of composite material to be processed can be deduced from Fig. 5. The analysis of Fig. 5 resulted in different evidence that could be useful to consider for CED minimization of a specific CFRTPs component. In detail, the PP-CF scenario is with a low performance polymer matrix and a high-performance reinforcement fibre. For the PP-CF scenario, the recycling and reforming EoL strategies are always comparable. Both processes are more advantageous than the combustion process. This advantage is even more evident as the percentage of reinforcement increases. For example, when considering the composite with 45% of reinforcement, the average values of the three EoL processes are 69.13 MJ, 13.99 MJ and 14.34 MJ, respectively. The PEEK-CF configuration is the one with the composite made of both high-performance polymeric matrix and fibre. The most competitive product's EoL is reforming, which has an energy impact that depends slightly on the fibre volume fraction percentage. The recycling process, instead, starts to become competitive just for the configuration with a high fibre percentage. Indeed, being the pyrolysis a recycling route that recovers the fibres, if the amount of reinforcement in the composite increases, the impact of the recycling process decreases. Conversely, if the matrix increases its prevalence, the pyrolysis wastes a higher quantity of high-performance matrix resulting in an increment of the energy impact close to the one ascribed to the combustion. Looking at the average values of the 3 processes, the values are respectively of 84.34, 30.89, 23.16 MJ for combustion, recycling, and reforming for a fibre percentage of 45%

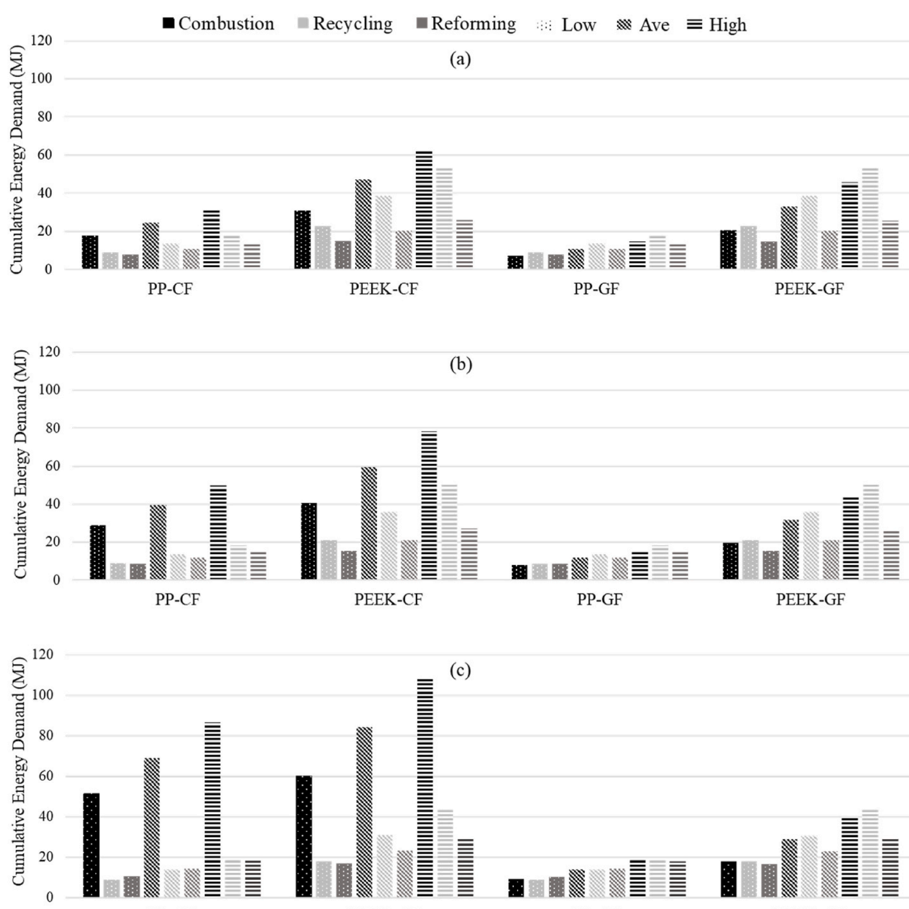

Fig. 5. CED required for each investigated FRPs and EoL routes at changing of (a) 11%, (b) 23%, (c) 45% FVF percentage.

while if this percentage passes to 11% the values change to 47.05, 38.56, 20.29 MJ, respectively. Looking at PP-GF, the composite material is made of both low-performance polymeric matrix and fibre. The EoL routes, i.e., combustion, recycling, and reforming processes, are comparable. The trend remains unchanged as the percentage of composite's reinforcement changes. Therefore, in this scenario, the combustion, being the simplest to be performed, is to be preferred, if CED is the index to be considered. Looking at the average values of the 3 processes, the values are respectively of 13.73, 13.83, 14.14 MJ for combustion, recycling, and reforming for a fibre percentage of 45% while if this percentage decreases to 11% the values remain comparable respectively to 10.83, 13.42, 10.77 MJ. The last consideration has to be, instead, reassessed, if specific midpoint or endpoint LCA indicators on use of raw materials are taken into account (Mio and Fermeglia, 2022). Considering the PEEK-GF, the composite material made from high-performance polymer matrix and low-performance fibre shows that the reforming process is the most promising process, especially if the configuration with low fibre volume percentage is taken into account. Furthermore, for this scenario, combustion is always preferred when compared to the recycling route. Looking at the average values of the 3 processes, the values are respectively of 28.90, 30.70, 22.88 MJ for combustion, recycling, and reforming for a fibre percentage of 45% while if this percentage decreases to 11% the values change respectively to 33.19, 38.52, 20.22 MJ.

Furthermore, the energy impacts of the different steps in the whole from-cradle-to-grave product's life were shown in Fig. 6, where the CED average values, for each considered FVF percentage, were reported. The weights of the recovered energies owing to the chosen EoL strategy are relevant, affecting the performances of the selected route, if products made by CFs are processed. Indeed, recovering CFs by recycling or reforming allows to reduce the energy impact of the products, markedly. For these configurations, reforming is, gradually, more promising than recycling at FVF's reduction if a performing polymer, i.e. PEEK, is processed. On the other side, if less valuable fibres are treated, i.e., GFs, the

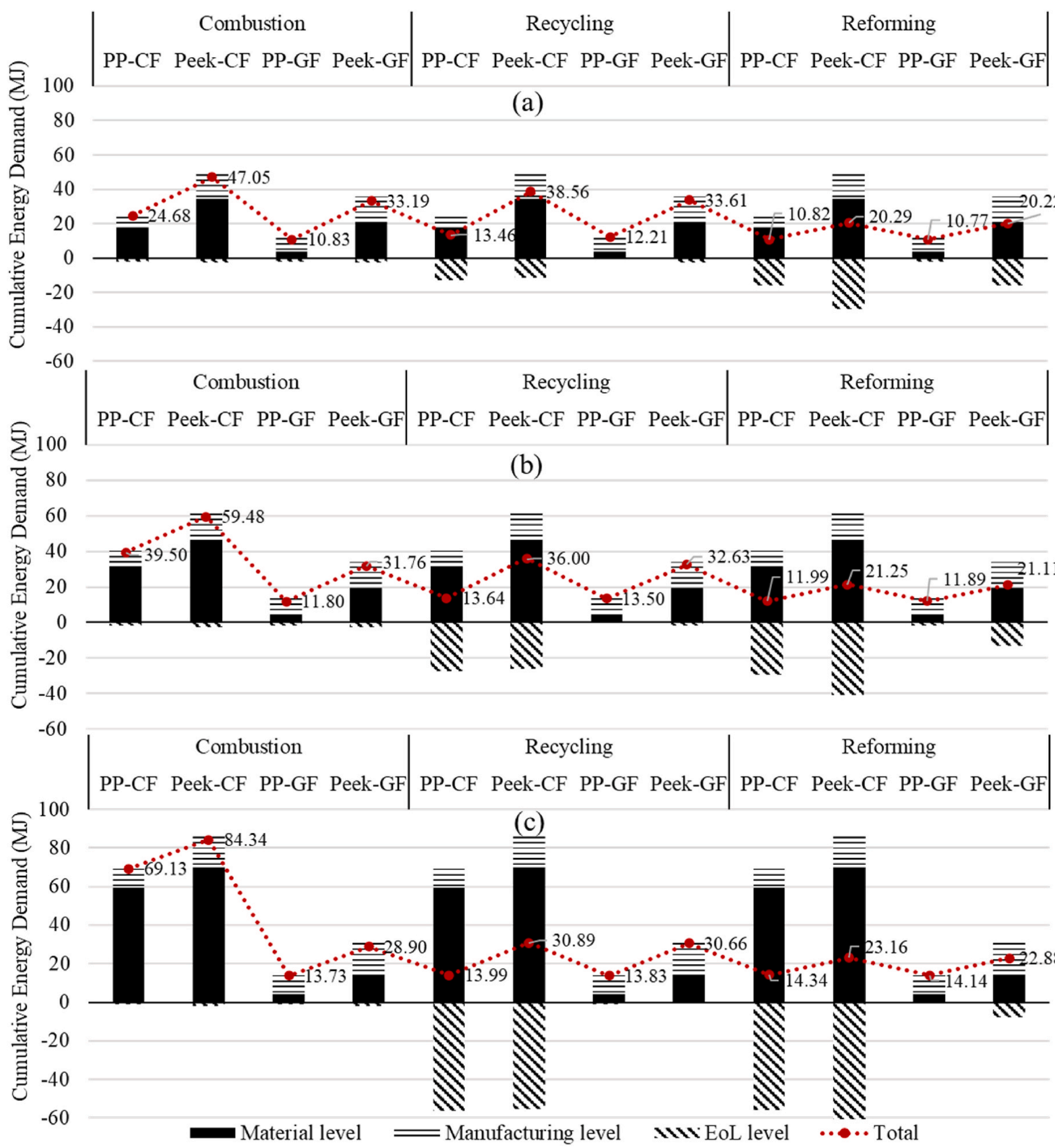

Fig. 6. energy impacts of the different steps in the whole from-cradle-to-grave product's life for (a) 11%, (b) 23%, (c) 45% of the analysed FVF percentage.

EoL routes, lose their weight on CED of the composite products, especially if GFs are combined to poor matrices.

5. Conclusions and outlook

A CED analysis was carried out by evaluating a target component made by the combination of two types of fibres, GF and CF, and matrices, PP and PEEK and considering also three FVF (11%, 23% and 45%). A life cycle energy analysis was performed evaluating three EoL routes. Specifically, applying the recycling content method approach, the CED of a specific product's life cycle was evaluated by using the data collected by a literature review. From this point of view, the possible EoL strategies were identified modelling, theoretically, the processing steps and gathering the information required for a proper CED quantification for each of them. The achieved LCI data are characterised by a huge dispersion in terms of embodied energies and energy consumption for both materials and manufacturing processes. In this respect, low, average and high values were reported, and used in the analysis to evaluate the products' CED with different material combinations.

The obtained results highlighted how, in a perspective of reducing the energy impact, the reforming EoL strategy is always a valuable solution to be taken into account. The choice can be more or less convenient depending on the type of composite processed looking at the utilised fibres and matrices and at the percentage of their employment in the composite construction. This research aimed at providing guidance for the selection of the most suitable EoL strategies, taking into account the CFRTPs material properties, as a decision support tool that, practically, can be employed in choosing the most energetically convenient path. The following recommendations can be extracted:

- if the composite is made of a low-value matrix and a high-value fibre, both reforming and recycling EoL routes can be used to minimise the energy impact;
- if the composite is made of a high value of both matrix and fibre, it is definitely worthwhile to use the reforming process, especially for low FVF of the FRPs;
- if the composite is made of a low value of both matrix and fibre, the energy impact of the three investigated EoL routes is comparable. Therefore, the choice of combustion, being the simplest solution, via incineration, should be preferable if CED is the only indicators to be considered;
- if the composite is made of a high-value matrix and a low-value fibre, reforming is more energetically convenient even if its advantages are more evident for low FVF;
- recycling is not always more advisable than combustion. Actually, if the composite is made by low-value fibres, combustion is to be preferred.
- the FVF can change the advantages of one EoL route with respect to another one. Specifically, particularly if high-value fibres are processed, recycling and reforming processes become increasingly comparable if the percentage of reinforcement increases and, at the same time, the combustion process becomes increasingly impactful.

As far as future developments are concerned, experimental tests need to be considered to assess the mechanical performance of the investigated composite product at different EoL routes. Loss in performance of the recycled fibres due to recycling phase or limitations in reforming owing to critical areas, such as bend angles and wrinkles onset, must be also considered, and evaluated as a future step for the development of the executed analysis. Furthermore, to be able to properly consider in the analysis also the solvolysis, namely the chemical recycling EoL route for CFRTPs, and its impacts due to the different solvents and catalysts used in recovering of both the polymer matrix and the full-length fibres, a LCA study in accordance with the ISO standards is required. By doing so, specific midpoint indicators, i.e., global warming, ionising radiation, and mineral resources, and endpoint indicators, i.e., human health, ecosystem quality and climate change, can be assessed to highlight the impacts of the investigated EoL solutions looking at their effects on different moments and environmental categories and providing a different point of view able to develop the results arisen by the CED analysis.

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Francesco Borda: Investigation, Conceptualization. **Giuseppe Ingarao:** Software, Methodology. **Giuseppina Ambrogio:** Formal analysis, Data curation. **Francesco Gagliardi:** Validation, Supervision, Conceptualization.

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.

Data availability

Data will be made available on request.

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