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Environmental Impact of an Innovative Aeronautic Carbon Composite Manufactured via Heated Vacuum-Assisted Resin Transfer Molding

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Abstract: The Vacuum-Assisted Resin Transfer Molding (VARTM) process has gained popularity as a reliable and cost-effective alternative to autoclave molding for high-performance composite production, which is especially interesting for aeronautics, where weight reduction is crucial. However, to date, the environmental impact of components produced through VARTM remains relatively unknown. To address this gap, this study applied the Life Cycle Assessment (LCA) methodology to estimate the environmental impact of a thermoset composite laminate produced through heated VARTM. Aiming to support the decision, the VARTM composite part's production was compared to conventional autoclave manufacturing, and the influence of alternative end-of-life (EoL) scenarios and energy mixes was considered, through LCA. The study found that energy consumption represented the majority of the environmental impacts of the heated VARTM component (33%), followed by carbon fibers, resins, consumables, and wastes. In terms of the comparative analysis, the autoclave manufacturing process showed better environmental results. Regarding EoL management, supercritical hydrolysis (with heat recovery) recycling emerges as the most beneficial method, reducing the impacts of the VARTM-manufactured component by 25%. This study emphasizes the importance of sustainable practices, such as reducing energy consumption, using low-carbon energy mixes, and adopting recycling methods to improve VARTM composite's environmental performance.

Keywords: vacuum-assisted resin transfer molding; carbon fibers; mono-component resin; life cycle assessment; aeronautics; recycling

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1. Introduction

Nowadays, nearly all aerostructures feature components fabricated from composite materials. Although composite materials were introduced in aeronautical structures in the 1950s, it is only recently that they have become a significant part of an aircraft, representing up to 50% of the Boeing 7877 Dreamliner [1]. With growing environmental concerns and the need to replace metal alloys currently used, composite materials, more specifically thermosets and thermoplastics, have been gaining more relevance and importance in the aeronautic industry since weight reduction can potentially reduce fuel consumption and therefore reduce operational emissions. These two composites vary based on their polymeric base. The key attribute distinguishing thermosetting composites is their resistance to softening or melting during heating cycles, characterized by complete cross-linking where polymer chains are interconnected through a network of bonds. In contrast, thermoplastic composites are capable of undergoing multiple heating and cooling cycles with minimal cross-linking, rendering them suitable for recycling as they can be reshaped and reformed repeatedly. However, given that the majority of composite structures in aeronautic applications are traditionally manufactured using autoclaves, and considering the high cycle times, capital investments, and tooling costs associated with autoclave processes, questions arise regarding the environmental and economic viability of composite materials [2]. Also, the dimensions of the autoclave can directly influence the production capacity, limiting the feasibility of the production of large components [3].

In this framework and considering the increases in demand and production of aircrafts, it becomes essential to develop and adopt solutions that are out of autoclave (OoA), aimed to be cheaper and hopefully more environmentally friendly. In the past, OoA solutions were mostly disregarded as the materials obtained were of inferior quality when compared to materials obtained via autoclaving. Currently, the new generation of OoA processes is gaining acceptance as they now offer the possibility to obtain composite materials with equivalent autoclave quality [4]. These processes use vacuum, pressure, and heat outside of the autoclave to manufacture composites. The most common OoA processes are vacuum bagging, oven cure, VARTM, quickstep curing, resin film infusion, and resin infusion under double-flexible tooling [5]. Of all of these processes, VARTM is one of the most robust and attractive replacements for autoclave processes [6]. VARTM is a closed-mold process that places dry reinforcements into a mold, seals them with a sealed flexible membrane (vacuum bag), and then, using differential pressure, infuses resin and impregnates the dry fibers. Depending on the chosen resin system, the process can be carried out at either ambient or elevated temperatures [7]. In contrast to autoclave processes, this method presents numerous advantages, including the ability to process cost-effective composites with complex geometries, ultimately yielding exceptional properties and a high-quality surface finish. It is also a closed process, resulting in minimal emissions of volatile organic compounds [8,9]. Moreover, VARTM significantly reduces tooling costs, making it the most cost-efficient technique available [10]. Although this method appears to offer many benefits, it also has some drawbacks. Skilled workers are necessary to operate it effectively, and long cycle times may result from extended mold-filling periods. Furthermore, consumables, such as sealing tape, peel ply, and vacuum bags, are required, and cannot be reused in most cases [11]. A further challenge is the complete impregnation of the dry fibers and the resulting porosity. Incomplete resin infusion leads to dry spots with a large number of voids, ultimately resulting in defective parts [12]. In response to this issue, a variant involving debulking the preform before the resin infusion has been developed. Unlike conventional VARTM, this debulking method utilizes two vacuum pumps, one at the inlet side and one at the outlet side, to reduce the pressure gradient in the preform. This reduction enhances the void content and ensures a more homogeneous thickness, though it may result in longer resin impregnation times [13].

Another challenge that has garnered attention in recent years concerns the end-of-life management of composite compounds. Actually, thermoset polymers make up approximately 15 to 20% of the plastics currently in production, with a predominant usage of carbon fibers strengthened by epoxy resins [14]. Predictions estimate that this number will continue to grow, so it is becoming increasingly urgent to find ways to recycle and reuse these materials. Regardless of their wide range of applications, thermoset polymers are difficult to recycle by the simple fact that these polymers are fully cross-linked and so, after polymerization, they cannot be melted and remolded. The absence of recyclability is not only an environmental setback, but also a limitation in potential applications and markets [15].

Currently, the main way of disposing of these components is through landfill or incineration. Landfilling, although the cheapest solution, does not allow the recovery of vast amounts of resources and it is also correlated to negative burdens. Through incineration, it is possible to recover energy; however, it is not an efficient process as it only recovers, in the form of heat, some of the embodied energy present in the thermosets and can produce secondary pollution [14,16]. These two traditional disposal solutions are not environmentally friendly, which has led, in recent years, to an increase in studies and incentives regarding the recycling of thermosets [17]. The technologies employed to recycle thermoset polymers can be divided into three categories: mechanical, thermal, and chemical. In mechanical recycling, the polymers are crushed or swelled to reduce their size and produce fine particles. Despite the fact that this process does not destroy the chemical structure of the polymer, it generates recycled products that normally have a low quality. Thermal recycling uses elevated temperatures to break down the polymers, separating the fibers from the thermoset resins. This process makes it possible to obtain clean fibers, although they have reduced mechanical properties. The volatilized organic compounds obtained from the resins are then used to produce energy, which helps offset the energy consumption of the entire process [18]. Chemical recycling aims to depolymerize the thermosetting composite by separating the fibers from the polymer matrix and decomposing the matrix into multiple chemicals, using dissolution reagents for this purpose. Fibers obtained through this method, when compared with fibers obtained by other methods, are generally cleaner and less degraded. However, the polymeric matrix is usually discarded and not recovered since the treatment conditions are mostly harsh, applying high temperatures and highly corrosive chemicals that are only intended to fully remove the resin and isolate and preserve the fibers. To optimize this recycling process, it is essential to recover both the fibers and the chemicals from the polymeric resin [19]. An effective approach that has been developed and implemented involves breaking down the polymeric matrix at mild conditions using substances such as hydrogen peroxide or acetic acid. This technique not only preserves the fibers, but also has the capacity to extract valuable chemical compounds, which can subsequently be repurposed into novel products [20].

Thermosetting composites have been increasing and may be dominant in future aeronautical structures. However, the OoA methods currently used to produce these structures are not fully optimized and the components obtained are not of the same quality as those obtained by autoclaving [4]. This study presents an innovative heated VARTM to process high-temperature mono-component epoxy resins, which, due to their high viscosity, cannot be used in conventional VARTM. With the current climate concerns of the aviation industry, it is important to fully understand the environmental impacts associated with the life cycle of these innovative materials. Yet, the environmental impact of components produced through VARTM remains relatively unknown. Thus, making use of the LCA methodology, this study aims to bridge this gap and evaluate the environmental performance of a thermoset composite laminate manufactured via heated VARTM by using carbon fibers and epoxy resin. The study provides primary life cycle inventory data, collected in situ, regarding the innovative VARTM panel production, for which literature data are still scarce, and compares the VARTM composite part to an autoclave-manufactured one. Considering the growing concern with the EoL of these composites, this study also models and assesses the environmental impacts of EoL recycling scenarios comparing them with the most common EoL solutions. Furthermore, a sensitivity analysis using alternative energy mixes is also presented.

2. Methodology

2.1. Goal and Scope

LCA is a methodology that systematically evaluates the potential environmental impacts of products, services, and materials from a life cycle perspective. It allows for the comparison of alternative systems that perform the same function. LCA is defined by the international standards ISO 14040:2006 [21], which defines the principles and structure, and ISO 14044:2006 [22], which addresses the requirements and guidelines. Following these standards, the LCA methodology should incorporate four interrelated phases: i. goal and scope, ii. life cycle inventory analysis (LCI), iii. life cycle impact assessment, and iv. results interpretation. This study applies the LCA methodology to assess the potential environmental impacts of a carbon fiber thermoset laminate manufactured via heated VARTM. The objective is to identify the key environmental hotspots of the product, and thereby facilitate decisions to improve the overall environmental performance. The LCA study is conducted using a "cradle-to-gate" approach for a functional unit of one aeronautic composite laminate.

Figure 1 presents the process flow chart and the system boundaries to produce one composite laminate via heated VARTM, which includes the following procedures:

- Fiber and technical fabrics cut—the first step is to cut the carbon fibers (20 layers) and the technical consumables (peel ply, flow mesh, spiral tubes, hoses, and vacuum bag).
- Mold, resin, and infusion preparation—the next phase begins with the removal of any impurities from the mold to ensure it is clean. Then the fibers and technical consumables are arranged in a predetermined orientation within the mold. Simultaneously, the resin is retrieved from the freezer and heated to 80 °C, the required temperature for degassing and removing all volatiles.
- Infusion—the infusion process starts with the fibers being impregnated with the resin. Prior to the infusion, the mold is heated, and a leak test and debulking process are conducted.
- Final steps—the resin undergoes curing at a temperature of 180 °C post-infusion. Subsequently, the resulting laminate is cut to yield a final product with dimensions of 460 × 260 mm.

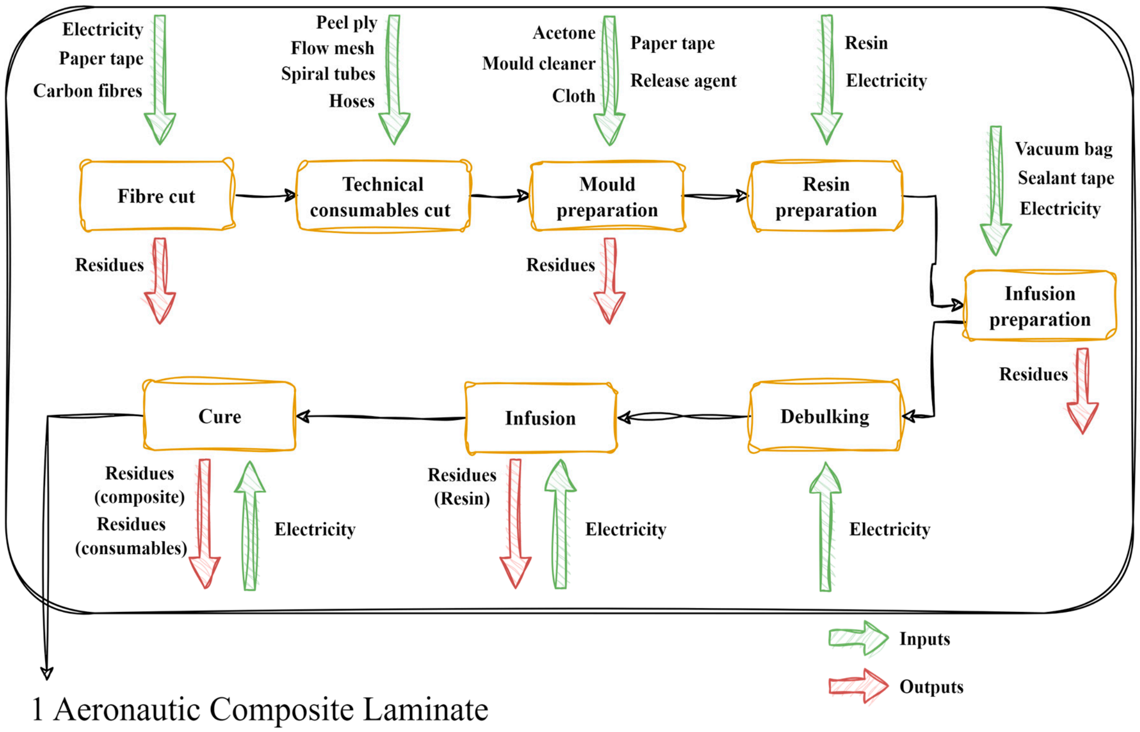

Figure 1. Flow chart and system boundary to produce one aeronautic composite laminate via heated VARTM.

2.2. Life Cycle Inventory

The LCI analysis comprises the collection and calculation of inventory data to quantify the input and output flows of the systems being studied.

2.2.1. VARTM Scenario

In this study, for the VARTM scenario, primary quantitative data were collected from the manufacturing processes based on real measurements at a pilot scale of production at INEGI lab facilities. Thus, the LCI data gathered, shown in Table 1, present novel quantitative data, which are mostly not yet available in the literature, documenting the VARTM production process for the functional unit. Primary information from the manufacturing process was complemented with environmental data from the EcoInvent database (v3.8) [23]. In cases where data were not available in EcoInvent, the literature was consulted to model proxies, as detailed below and in Table S1. These proxies serve as substitutes for missing inputs and outputs from the database, enabling the effective modeling of the environmental impacts of the process. To account for carbon fiber production, data from the study of Forcellese et al. [24] were used. Based on Hill's and Norton's report [25], the mold cleaner and the release agent were replaced with "methyl ethyl ketone" and "chemical, organic", respectively. The peel ply and flow mesh were replaced by "nylon 6-6" and "polypropylene", respectively, based on the study conducted by Gkoloni and Kostopoulos [26], while the hoses were replaced by "polyamide" according to La Rosa et al. [27]. Due to a lack of information in the literature, alternative materials were utilized as proxies for paper tape, spiral tubes, and vacuum bags, namely, "tissue paper" [28], "low-density polyethylene" [29], and "nylon 6-6" [30], respectively. These proxies were selected because they are the main components of their respective materials. Additionally, the total electricity energy consumption associated with the VARTM manufacturing process was inventoried based on local measurements. Therefore, the total electricity consumption was accounted for.

Table 1. LCI of the inputs and outputs to produce one aeronautic composite laminate, via heated VARTM.

Process Steps	Input	Quantity	Unit	Output	Quantity	Unit
Fiber cut	Carbon fibers	0.70	kg	Carbon fiber residues	0.12	kg
	Paper tape	0.02	kg	-	-	-
Technical consumables cut	Peel ply	0.014	kg	-	-	-
	Flow mesh	0.013	kg	-	-	-
	Spiral tubes	0.044	kg	-	-	-
	Hoses	0.23	kg	-	-	-
Mold preparation	Acetone	0.015	kg	Consumables residues	0.14	kg
	Mold cleaner	0.047	kg	-	-	-
	Release agent	0.026	kg	-	-	-
	Cloth	0.045	kg	-	-	-
	Paper tape	0.010	kg	-	-	-
Resin preparation	Resin	1.3	kg	-	-	-
Infusion preparation	Vacuum bag	0.025	kg	Consumables residues	0.021	kg
	Sealant tape	0.28	kg	-	-	-
Debulking	-	-	-	-	-	-
Infusion	-	-	-	Resin residues	1.1	kg
Cure cycle	-	-	-	Laminate residues	0.14	kg
	-	-	-	Consumables residues	0.60	kg
Energy consumption	Electricity	36	kWh	-	-	-

2.2.2. Autoclave Scenario

In order to evaluate whether the VARTM manufacturing process is more advantageous than the traditional autoclave manufacturing process, a comparison was made using the same functional unit. Table 2 presents the required inputs and outputs along the different process steps for the autoclave process. Most details related to autoclave manufacturing were obtained through empirical measurements, while the remaining data were estimated based on proxies from the literature. The study conducted by Katsiropoulos et al. [31] was used to determine the amount of electrical energy consumed. To determine the consumables used, a combination of mass allocations based on empirical data and research conducted by the study by Forcellese et al. [24] was used.

Table 2. LCI of the input and outputs to produce one aeronautic composite laminate, via autoclave.

Process Steps	Input	Quantity	Unit	Output	Quantity	Unit
Cut and stack prepreg	Thermoset prepreg	0.81	kg	-	-	-
Autoclave preparation	Peel ply	0.015	kg	-	-	-
	Release film	0.047	kg	-	-	-
	Breather	0.045	kg	-	-	-
	Blue tape	9.0×10^{-3}	kg	-	-	-
	Sealant tape	0.58	kg	-	-	-
	Vacuum bag	0.026	kg	-	-	-
Autoclave	-	-	kWh	Consumables residues	0.13	kg
Final cut	-	-	kWh	Laminate residues	0.14	kg
Energy consumption	Electricity	4.0	kWh	-	-	-

2.2.3. EoL Scenarios

To address the problem of composite component disposal, this study examined three recycling scenarios and compared them to two conventional disposal methods. One of the conventional options analyzed involved sending the composite to a landfill, while the other involved sending it to a combination of plastic treatments, including incineration, open burning, and landfill. The study examined three recycling scenarios: one involving chemical recycling under mild conditions, another involving chemical recycling under supercritical conditions, and a third involving chemical recycling under supercritical conditions with waste energy recovery. The purpose of these scenarios was to recover and reuse the carbon fibers from the composites. Chemical recycling was preferred over other recycling methods as it typically results in less degraded and cleaner fibers, while also breaking down the resin into valuable chemicals [32]. In this study, the recycled fibers were assumed to have a similar quality to virgin fibers and could be further used to replace virgin ones, whereas the chemicals obtained from the resin were not utilized and were still treated as waste. It is also important to note that the fiber/resin ratio utilized in the present study differed from ratios used in the studies where the inventories were retrieved, emphasizing the need for a discerning approach. The fiber-to-resin ratio is a significant factor that varies depending on the intended composite and the type of resin used. These differences in ratios can have an impact on the yield of the recycling process and the environmental impacts of the composite.

As reported in the La Rosa et al.'s study [32], the use of mild conditions in the chemical recycling of composites is one of the most used methods for separating carbon fibers from resin. In that study, the composite was treated in a solution of acetic acid at a temperature of 80 °C for 1.5 h. After treatment, the mixture was filtered, allowing the carbon fibers to be separated from the remaining chemicals. To neutralize the acetic acid, hydroxy sodium was added until a solid appeared. The mixture was then washed with water at 40 °C. By adding a few drops of sodium hydroxide to the mixture, an epoxy thermoplastic polymer was precipitated. The process is depicted in Figure 2a. According to the literature [32], a significant proportion of the carbon fibers utilized in the production of the composite material can be reclaimed, with a recovery rate of up to 94%.

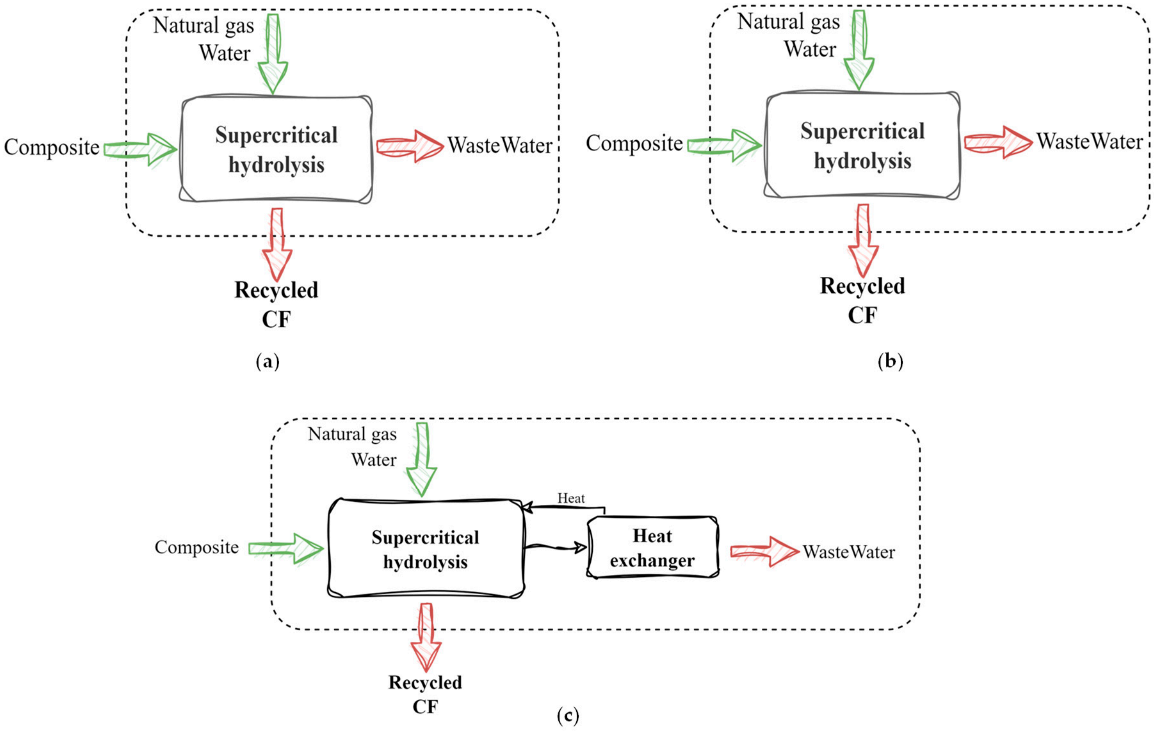

Figure 2. Flow chart and system boundary for (**a**) mild hydrolysis; (**b**) supercritical hydrolysis; and (**c**) supercritical hydrolysis with waste heat recovery (WHR).

Supercritical hydrolysis is considered a promising approach for recycling composites, although it is still in the experimental stage and requires further refinement at the laboratory level [33]. This approach utilizes a fluid that is at its critical temperature and pressure, which exhibits excellent solubility and diffusivity, to disintegrate the composite, as described in Figure 2b. Water and alcohol are the most commonly used solvents for this chemical process, either with or without the addition of catalysts. Unlike many recycling processes, this particular method does not require any pre-treatment. The composite can be directly placed into the reactor, which should be appropriately sized to handle both the composite and the desired fibers. According to Pillain et al. [33], the supercritical hydrolysis process begins by heating water to 374 °C (which is the critical temperature of water) and pressurizing it to 25 MPa. The heated and pressurized water is then introduced into the reactor, causing the polymer matrix to undergo hydrolysis and the resin to break down. The resulting outcome would be clean and isolated fibers, with the research presuming the complete recovery of the fibers. Also, in Pillain et al.'s study, an improved method was proposed, which involved the installation of a heat exchanger. This allowed for the recovery of heat from the output stream, which could then be used to pre-heat the input water, as shown in Figure 2c. Our study assessed the impacts of heat recovery on overall recycling performance.

A full life cycle inventory data table for the EOL scenarios is presented in the Supplementary Information in Tables S2–S4.

The LCI data for the inputs and outputs of the recycling processes were obtained from La Rosa et al.'s [32] and Pillain et al.'s [33] studies and complemented by additional information from the EcoInvent database. It was considered a distance of 9 km, which represents the distance between the manufacturing location and the nearby recycling site. It was also assumed that the quality of the recovered fibers is equivalent to that of virgin fibers and that they can be reintroduced into the system as replacements. The recycling models were developed based on one kilogram of the composite material and then extrapolated to the same functional unit of the VARTM scenario.

2.2.4. Sensitivity Analysis

For a more comprehensive assessment of the environmental impact associated with an aeronautical composite laminate manufactured via heated VARTM, and to analyze the influence of energy mixes, this study performed a sensitivity analysis. The assessment was conducted with reference to three energy mix scenarios: the Portuguese energy mix, serving as the baseline; the Norwegian energy mix, characterized by a higher proportion of renewable energy sources; and the German energy mix, which primarily relies on fossil fuels with a limited incorporation of renewable sources. The data utilized for these scenarios were sourced from the EcoInvent database (v3.8), as detailed in Table 3.

Table 3. Comparison of energy mixes in Portugal, Norway, and Germany.

	Renewable	**Fossil**	**Nuclear**	**Cogeneration**	**Others**
Portugal	47.4%	40.7%	-	1.6%	10.3%
Norway	94.4%	0.02%	-	0.1%	5.4%
Germany	25%	39.4%	13.5%	16.3%	5.7%

2.3. Life Cycle Impact Assessment

During the Life Cycle Impact Assessment (LCIA) stage the "CML-IA baseline V3.08/ EU25" method and SimaProTM 9.3.0.3 software were used to estimate the environmental impacts. Table 4 provides a complete list of the CML midpoint impact categories that were analyzed.

Table 4. Impact categories.

Categories	**Abbreviation**	**Unit**
Abiotic Depletion Potential (elements)	ADP elements	kg Sb eq
Abiotic Depletion Potential (fossil)	ADP fossil	MJ
Global Warming Potential	GWP	kg CO_2 eq
Ozone-Layer Depletion Potential	ODP	kg CFC-11 eq
Human Toxicity Potential	HTP	kg 1,4-DB eq
Freshwater Aquatic Ecotoxicity Potential	FAETP	kg 1,4-DB eq
Marine Aquatic Ecotoxicity Potential	MAETP	kg 1,4-DB eq
Terrestrial Ecotoxicity Potential	TETP	kg 1,4-DB eq
Photochemical Ozone Creation Potential	POCP	kg C_2H_4 eq
Acidification Potential	AP	kg SO_2 eq
Eutrophication Potential	EP	kg ${PO_4}^{3-}$ eq

3. Results

3.1. VARTM Scenario

The quantitative environmental impacts related to manufacturing one aeronautical composite laminate by VARTM are presented in Table 5. A detailed table of the impacts by process step is provided in Table S5. Figure 3 illustrates the relative impact of each process phase.

Figure 3 depicts the main environmental impacts of the vacuum infusion process, which are mainly attributed to electricity consumption, accounting for an average of 33% of the total environmental impacts. The amount of electricity consumed is linked to the temperature levels required for heating the resin up to 80 °C, as well as the high-temperature ranges needed for the injection and curing stages. In comparison with the findings in the literature, processes like resin transfer molding and vacuum infusion, which rely most on pressure and vacuum and do not subject the resin to such high temperatures, do not consume as much energy as the process developed in this study [2,34]. The cutting of fibers and preparation of resin phases are also significant hotspots, being responsible for 28% and 23% of the total impacts, respectively. The environmental impacts coming from the fiber cutting step predominantly arise from the utilization of carbon fibers and their production, which is highly energy intensive, and the emissions released during the production of the synthetic precursor, polyacrylonitrile [35]. This finding aligns with the existing literature, where OoA studies assert that materials containing carbon fibers are hotspots in the process, exhibiting significant environmental footprints [2,24,34]. On the other hand, the environmental impacts associated with resin preparation are primarily due to the large quantities used in the process, considering the relatively small size of the laminate produced. At the laboratory scale, a significant amount of resin remains in the pipes compared to the actual amount applied to the fibers. With an increase in the production scale, this issue is anticipated to be improved.

Table 5. Midpoint impacts for the production of one aeronautic composite laminate.

Categories	Unit	Impacts
ADP elements	kg Sb eq	1.40×10^{-4}
ADP fossil	MJ	434
GWP	kg CO_2 eq	31.3
ODP	kg CFC-11 eq	2.25×10^{-6}
HTP	kg 1,4-DB eq	19.5
FAETP	kg 1,4-DB eq	15.0
MAETP	kg 1,4-DB eq	3.45×10^{4}
TETP	kg 1,4-DB eq	0.109
POCP	kg C_2H_4 eq	1.46×10^{-2}
AP	kg SO_2 eq	0.176
EP	kg PO_4^{3-} eq	0.406

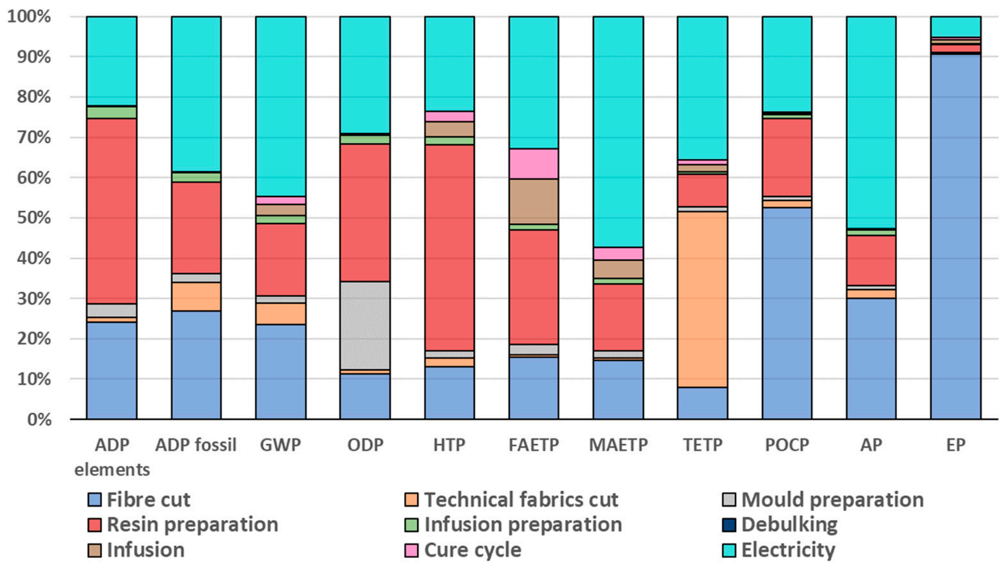

Figure 3. Relative environmental impacts associated with the production of one aeronautic composite laminate by heated VARTM.

Although not considered hotspots, the use of consumables and the generation of waste can have a significant impact on the VARTM manufacturing process. Despite the fact the overall environmental impact of these consumables and wastes is not clearly visible in Figure 2, due to their dispersion throughout various process steps, they still represent a notable share of the overall impacts, accounting for 11% and 6% of the total impacts, respectively, as it is possible to see in Figure 4a. Figure 4b reveals that hoses, sealant tape, and cloth are the main sources of the environmental impacts (34%, 21%, and 20%, respectively) associated with the consumables utilized in the VARTM manufacturing process. Comparable to other consumables, those with the highest share of impacts are the ones that are used in larger quantities. These include materials that have compounds such as polyethene and polyester.

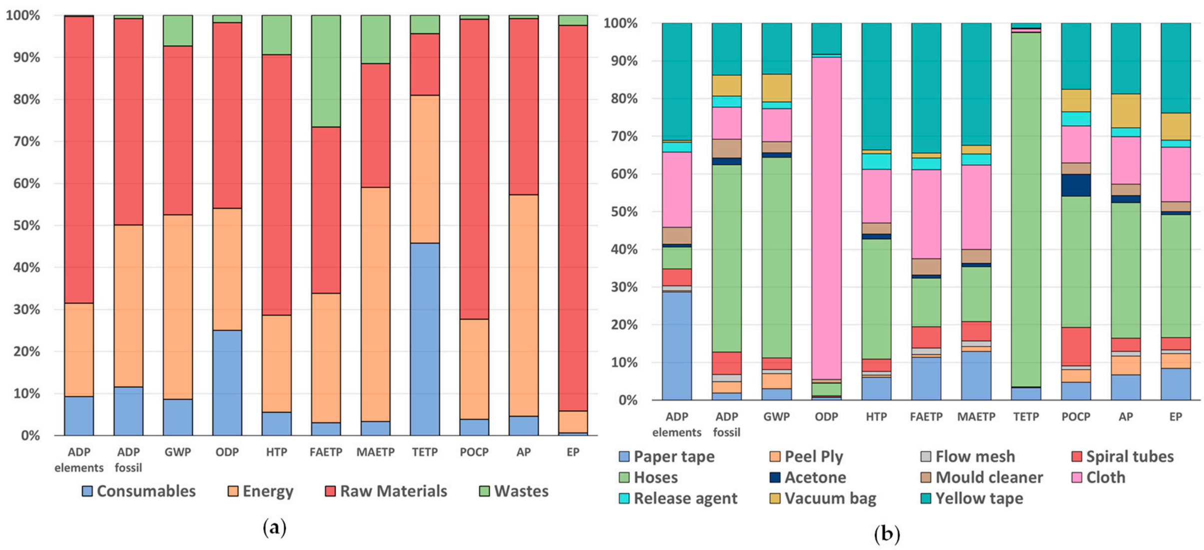

Figure 4. (**a**) Relative contribution of inputs and outputs and (**b**) relative contribution of the consumables.

The impact of wastes is primarily determined by their disposal methods. In this case, the residues generated are mostly sent to waste plastic treatment facilities where they may be disposed of through landfilling, incineration, or open burning. Despite the small quantity of waste generated, these disposal methods are highly detrimental to the environment, justifying the significant share of impacts.

3.2. Autoclave Scenario

In the autoclave scenario, as illustrated in Figure 5a and outlined in Table S6, the cutting of prepreg materials is identified as the primary contributor to environmental impacts, accounting for an average of 60% of the total impact. These impacts are mostly attributed to the use of carbon fibers and resin in prepreg manufacturing, as was also observed in the VARTM process, rather than the specific cutting process itself. The second most significant impact source, accounting for 28% of the total impacts, is the autoclave preparation phase. The impacts of this phase are mainly associated with the consumables used, which, as demonstrated in the VARTM scenario, can also lead to significant impacts. Electricity consumption is the main difference between the heated VARTM manufacturing process, where electrical consumption represents 33% of the total impact, and the autoclave manufacturing process, where it accounts for 9% of the total impact. While it is estimated that the autoclave process requires around 4 kWh, the VARTM manufacturing process requires 36 kWh. This difference in electrical consumption has a significant influence on the overall impacts of both processes, as shown in Figure 5b, with the autoclave generating 57% less impact than the vacuum infusion process. This result is opposed to what is typically found in the available literature, where the autoclave usually presents a high energy consumption rate, which translates to poorer environmental results compared to other OoA processes [2,31].

To better understand these comparative results, one must consider the size and capacity of the equipment used, as well as the novelty of the process being developed. Autoclave equipment has the advantage of being able to process multiple laminates at once, resulting in the distribution of the energy consumed among the components being processed. If a full production capacity is used for each curing cycle, this distribution can lead to a lower environmental impact per part being molded. Whereas, in the analyzed heated VARTM manufacturing process, significant heating-up energy and resin are lost each time a part is manufactured since the process is not yet optimized. The difference in capacity and efficiency may justify why the autoclave, despite being known for its high energy intensity, in this case displays a lower environmental impact.

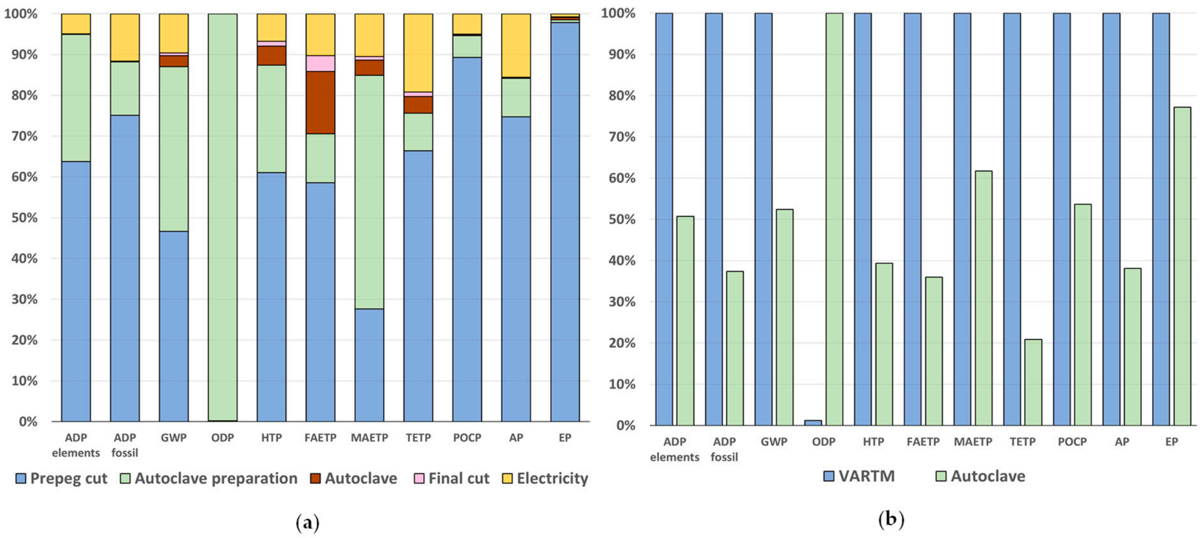

Figure 5. (**a**) Relative environmental impacts associated with the production of one aeronautic composite laminate using the autoclave process and thermoset prepregs and (**b**) comparison between heated VARTM and autoclave processes.

For the small lab batch analyzed, the most significant differences can be seen in the TETP category, where the impact is 79% lower for the autoclave manufacturing process. The ODP category is the only one where the autoclave process has a higher impact than the VARTM process. The increased environmental impacts observed in the ODP category for the autoclave method can be linked to the utilization of a release film, which is absent from the VARTM manufacturing process and has a higher impact on this category than other consumables. This outcome is primarily due to the precursor of the release film, named chlorodifluoromethane, which has a significant impact on the ozone layer [36]. It is important to note that, in a higher batch or continuous production, less resin and energy may be required per part by VARTM, which may shift the results of this comparative analysis. Thus, further studies, based on primary LCI documenting the production of bigger batches (or production scales) are advisable. Another option to consider could be using VARTM manufacturing processes operating at lower temperatures or with shorter curing times. From an environmental point of view, improvements regarding the processing capacity and its energy efficiency are required since energy consumption is a hotspot that may hinder the environmental performance of the VARTM-manufactured parts.

3.3. EoL Scenarios

The environmental impacts for the recycling scenarios chosen, mild hydrolysis, supercritical hydrolysis, and supercritical hydrolysis with a heat exchanger, are shown in Figure 6a–c, respectively. The quantitative values are further detailed in Table S7. The primary cause of concern in the mild hydrolysis recycling process is the production of the acetic acid required for hydrolysis, which accounts for approximately 49% of the overall environmental impact of the process. Additionally, the energy required to heat the solutions also has a significant impact, contributing to an average of 30% of the total environmental impact. For the supercritical hydrolysis scenario, it is clear to observe that the main hotspot is the natural gas required to heat and pressurize the water to its supercritical point, accounting for 84% of the environmental impacts. However, the incorporation of a heat exchanger into the process has the potential to significantly diminish the importance of natural gas. In this particular scenario, natural gas contributes only 50% of the total impacts. The incorporation of a heat exchanger enables the process to reuse heat, resulting in an overall reduction in impacts of approximately 75%. On the other hand, while the amount of water consumed remains equal, the relative importance of wastewater increases. Prior to the installation of the heat exchanger, wastewater represented only 13% of the overall impacts, but now it accounts for 38% of the total impacts. The high percentage is mainly attributed to the large quantities of water required for the supercritical hydrolysis process.

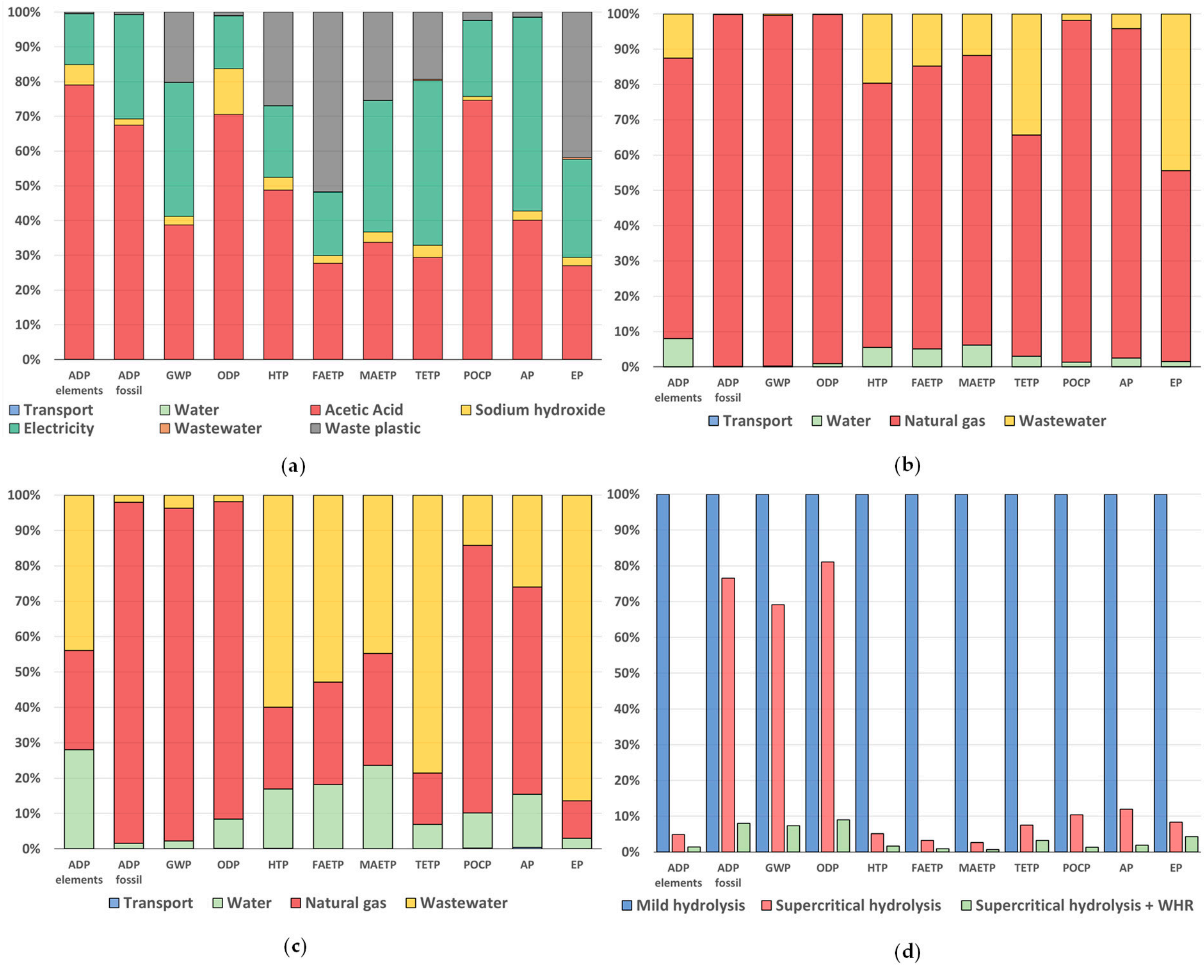

Figure 6. Relative environmental impacts for the (**a**) mild hydrolysis scenario, (**b**) supercritical hydrolysis scenario, and (**c**) supercritical hydrolysis incorporated with a heat exchanger; (**d**) comparison between mild hydrolysis, supercritical hydrolysis, and supercritical hydrolysis with waste heat recovery (WHR).

In Figure 6d, a comparison of the recycling scenarios shows that supercritical hydrolysis yields significantly better results than mild hydrolysis, with the impacts being on average 74% lower. Moreover, the inclusion of a heat exchanger in the supercritical hydrolysis scenario results in a 96% reduction in impacts compared to mild hydrolysis. The primary difference between mild and supercritical hydrolysis lies in the solvent used in each process, with acetic acid being much more impactful than water.

The recycling processes depicted have varying impacts that influence the overall environmental effects of aeronautical component production. In the case of the laminate recycled under mild recycling, EoL impacts make up 4% of the total environmental impact while recycling with supercritical conditions and the integration of a heat exchanger contributes just 0.2%.

From Figure 7, it is possible to observe that the implementation of these recycling methods, with the respective avoided burdens (due to future carbon fiber reuse) may have a positive impact on reducing the environmental impacts associated with the aeronautic component production process. However, when comparing both scenarios, it is important to consider the respective yields. The mild hydrolysis process, with a yield of 94%, can recover 0.45 kg of fibers from the final laminate. In contrast, the supercritical hydrolysis process has a 100% yield and can recover 0.48 kg. The figure illustrates that recycling and reusing carbon fibers through the mild hydrolysis process can lead to environmental impact reductions ranging from 4% to 58%, with the POCP and EP categories experiencing the most significant reductions of 33% and 58%, respectively. Meanwhile, the supercritical hydrolysis process, with heat recovery, achieves reductions ranging from 5% to 62%, with the POCP and EP categories once again having the most substantial reductions of 36% and 62%, respectively. These categories are primarily associated with the emissions generated during carbon fiber production.

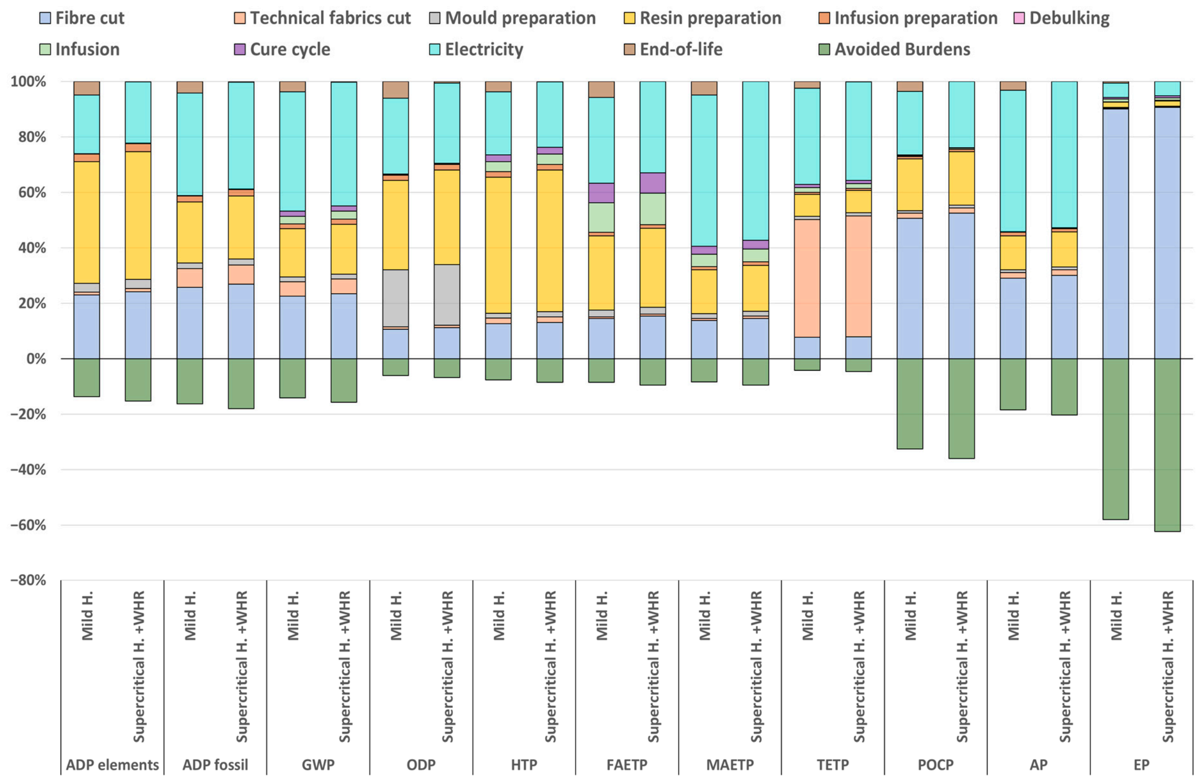

Figure 7. Relative environmental impacts associated with the production of one aeronautic composite laminate by heated VARTM, with avoided burdens (Note: H. stands for hydrolysis and WHR stands for waste heat recovery).

The avoided burdens that can be achieved through material reuse and the energy-intensive nature of carbon fiber production make recycling an appealing waste treatment option. Nevertheless, to assess the environmental benefits, it is essential to compare the environmental impacts of recycling methods to conventional disposal methods. For this purpose, a comparison was conducted to evaluate the impacts of two conventional disposal methods with the two recycling methods. Notably, the total impacts of the process were not significantly affected by the two conventional disposal methods, with only 3% attributable to landfill and 1% to the mixed treatment of plastics. However, from Figure 8, it is evident that conventional disposal methods result in higher environmental impacts compared to recycling methods, especially landfill. The main reason for the higher environmental impacts of conventional disposal methods is that, unlike recycling methods, they deter material recovery. This results in a significant amount of waste that cannot be reused, leading to higher impacts. When comparing landfill disposal, the worst-case scenario, with recycling methods, we observed that the mild hydrolysis scenario decreased the impacts by 16%, while the supercritical hydrolysis scenario reduced them by 21%.

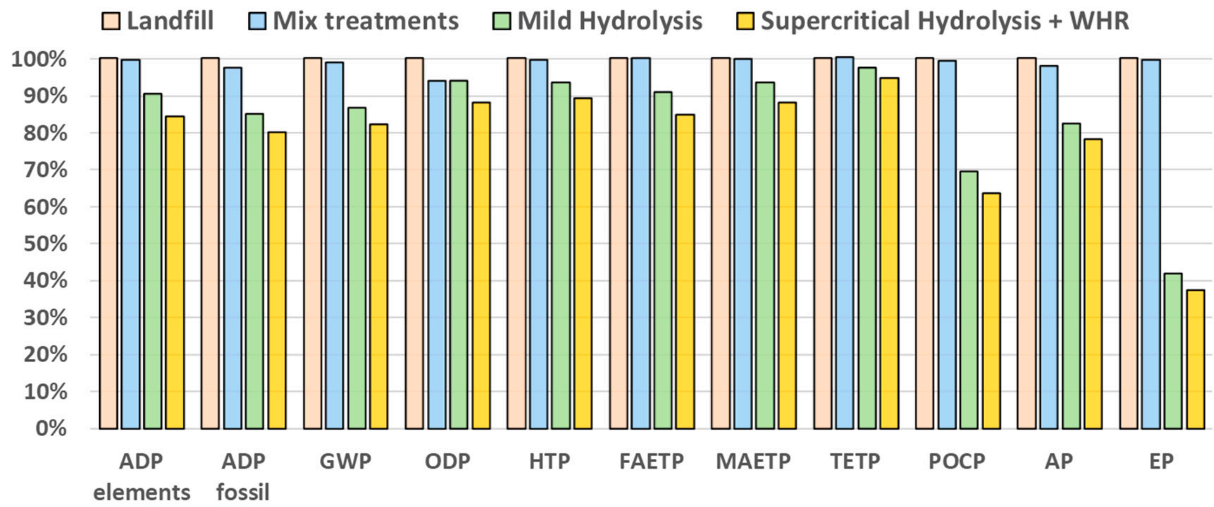

Figure 8. Relative environmental impacts associated with the production of one aeronautic composite laminate by heated VARTM, comparing non-recycling scenarios (landfill and plastic mix) with recycling scenarios: mild and supercritical hydrolysis with waste heat recovery (WHR).

3.4. Sensitivity Analysis

As demonstrated before, the VARTM process consumes a significant amount of electricity. One solution to this issue is to optimize the process and equipment, while another is to switch to a more sustainable energy source. This study examined the effects of changing the energy mix and the importance of renewable energy sources in reducing the environmental impact of energy production.

The analysis revealed that, when the Portuguese energy mix was used, electrical consumption contributed to 33% of the total impacts of the VARTM manufacturing process. However, when the German energy mix was used instead, the environmental impacts increased, with electrical consumption accounting for 42% of the total impacts. Conversely, using the Norwegian energy mix resulted in a significant decrease, with electrical consumption contributing only 8% of the total impact.

As shown in Figure 9, the use of the German energy mix results in greater environmental impacts compared to the scenarios involving the Portuguese and Norwegian energy mixes. Despite Germany's mix being more diverse, it has the lowest proportion of renewable sources among the three. The Portuguese scenario, although having a significant proportion of fossil fuels, features a noteworthy share of renewable energy, particularly wind energy. When compared to the German energy mix, utilizing the Portuguese mix can result in savings of up to 23% in terms of environmental impact. The only categories where the Portuguese mix has a greater impact than the German mix are the POCP and AP categories, primarily due to the use of hard coal, which is 76% higher in the Portuguese mix than that used in the German mix. Out of the three scenarios, the Norwegian mix stands out as having the most favorable results. With a remarkable 94.4% share of renewable sources, it comes as no surprise that it exhibits the lowest environmental impacts. In comparison to the Portuguese mix, using the Norwegian mix could result in savings of up to 23% of impacts. These results highlight the importance of using renewable energy sources to reduce the environmental impact of highly energetic processes.

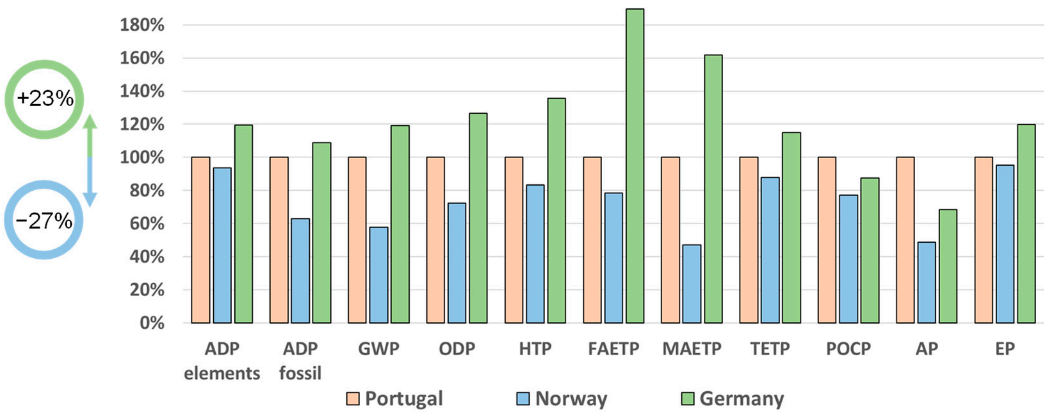

Figure 9. Sensitivity analysis and impacts associated with the production of one aeronautic composite laminate heated VARTM using energy mixes from Portugal, Norway, and Germany.

4. Conclusions

The life cycle environmental impacts of manufacturing an aeronautical composite laminate by heated VARTM were investigated in this study, using the LCA methodology. A comparison of the heated VARTM manufacturing process with the conventional autoclave manufacturing process is presented based on laboratory production LCI and literature data. The results obtained for the heated VARTM reveal that the manufacturing process's primary environmental impact occurs due to electricity consumption, accounting for 33% of the total impact. There is an additional concern regarding carbon fibers and resin., along with consumables required and waste generated during the process. The autoclave process showed better environmental results than the heated VARTM manufacturing process that was developed, with a 57% reduction in environmental impact. The primary factor contributing to this significant environmental contrast between the two methods was their disparity in energy consumption. However, it is important to note that interpreting the results requires a careful consideration of the underlying assumptions made in the study. The autoclave, despite being performed at the lab scale, was considered to simultaneously cure a batch of components, whereas the lab-heated VARTM could only cure a part at a time. Since the process has not yet been optimized, significant resin and heat are lost during the production of each part. Thus, further research is recommended to be carried out for higher production batches.

The study also addressed the issue of composite EoL disposal, which typically involves landfills or incineration. To comprehend the potential benefits of recycling methods, this study compared mild hydrolysis and supercritical hydrolysis (with and without waste heat recovery) with conventional EoL methods (landfill and mixed-plastic treatment). The results show that supercritical hydrolysis has the lowest environmental impact. Compared to landfill, the mild hydrolysis method reduced impacts by 19%, while the supercritical hydrolysis with the waste heat recovery method reduced impacts by 25%.

Additionally, a sensitivity analysis was conducted to better understand the heated VARTM's environmental profile. The analysis involved replacing the Portuguese energy mix (used in the baseline scenario) with the Norwegian and German mixes. The results show that the German mix has the highest environmental impact, with the Portuguese mix presenting savings of up to 23% when compared to the former. Notably, the Norwegian mix, characterized by a significant share of renewable energy, demonstrated even better results, achieving savings of up to 27% compared to the Portuguese mix.

This study, based on lab experimental processes, provides insights into the environmental impacts of aeronautical composite laminate manufacturing using the heated VARTM manufacturing process. The study's findings suggest that the supercritical hydrolysis recycling method offers a better environmental performance than the other methods analyzed in the study.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su16083253/s1, Table S1. Proxies and references used to model the life cycle inventory (LCI) data (inputs and outputs) for the different scenarios. Table S2. LCI of the inputs and outputs to process 1 kg of residues by Mild hydrolysis. Table S3. LCI of the inputs and outputs to process 1 kg of residues by Supercritical hydrolysis. Table S4. LCI of the inputs and outputs to process 1 kg of residues by Supercritical hydrolysis with heat recovery. Table S5. Midpoint impacts for the production of one aeronautic composite laminate via VARTM, by phase of production. Table S6. Midpoint impacts for the production of one aeronautic composite laminate via autoclave, by phase of production. Table S7. Midpoint impacts for the EOL scenarios. References [26–32] are cited in the Supplementary Materials.

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