

Reuse of Fiber Reinforced Polymer Waste in MSLA 3D Printing: Mechanical Properties and Sustainability Assessment

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Abstract. The use of composite materials and specifically of Fiber Reinforced Polymers (FRP) is continuously increasing in structural applications due to their high strength-to-weight ratio. From an environmental perspective, composites still face relevant challenges due to impactful petroleum-based matrices and large amounts of waste generated during manufacturing processes. This study proposes the reuse of FRP machining waste as filler in Masked Stereolithography (M-SLA) 3D printing. Scraps from FRP laminates, obtained during drilling operations, were incorporated into a photocurable resin and used to print tensile and flexural specimens with increasing filler contents (0–5 wt%) and mechanical characterization tests were carried out. A cradle-to-grave Life Cycle Assessment (LCA) was performed to quantify the potential environmental benefits associated with the reduced use of virgin resin. Results show that the use of recycled FRP waste leads to a loss of tensile strength and stiffness (up to 61% and 21% respectively) but it also provides a reduction in Global Warming Potential (about 2% at 5 wt% filler). This demonstrates that the proposed strategy can improve the sustainability of 3D-printed components, especially for non-structural applications.

1. Introduction

The use of Fiber Reinforced Polymers (FRP) in the market has significantly grown in the last decades due to their high mechanical properties, lightness and tailorable physical properties. These characteristics make composites an ideal solution for high performance sectors such as aerospace, automotive, maritime and energy [1]. However, their widespread use presents some sustainability challenges, in particular because of the impacts related to raw materials extraction, waste produced during the manufacturing processes and the difficult End of Life (EoL) management. In particular, this latter aspect is especially critical for thermoset polymer matrix composites, where recycling is particularly challenging. Many recycling technologies have been developed, and they are based on mechanical, thermal or chemical processes [2]; many of these methods remain energy-intensive and expensive, with limited adoption in industry. In addition, the recovered materials have typically degraded properties with respect to the virgin composites.

Filament Winding (FW) is a highly automated manufacturing processes for FRP used for axisymmetric components such as pipes and pressure vessels. FW is based on the deposition of impregnated fibers tows on a rotating mandrel via a computer-controlled machine, allowing to produce high performance structures. Despite its advantages, FW can be associated with relatively high energy consumption and may still generate composite waste, in particular during post processing operations such as drilling and trimming (i.e. of outer regions of the manufactured components).

Simultaneously, 3D printing is a promising manufacturing technology due to the possibility of producing complex geometries, reducing material use and waste. These technologies allow to produce components layer by layer, positioning material only where needed according to precise CAD models. Amongst the possible 3D printing technologies, Stereolithography (SLA) is one of the most commonly used; it is based on the use of photopolimeric resin that harden due to the effect of a light source (e.g. laser, LCD monitor...). Also in the case of 3D printing, despite the possible advantages in terms of

weight reduction and optimized processes, some issues related to environmental sustainability still arise. In fact, most of the virgin material used for these processes (resin, filaments...) have a synthetic origin (i.e. petroleum based) and are therefore associated with relevant environmental impacts. Hence, material use reduction and parts weight optimization is a crucial aspect in 3D printing to limit the final components environmental impacts.

In this context, photopolymer resins are often added with particulate or fiber fillers to enhance specific mechanical or physical properties or to reduce overall resin consumption. Scientific literature provides numerous examples of these solutions. Mineral and ceramic such as silica, alumina and talc were added to the resin with possible benefits in terms of increase in strength, modulus and wear resistance [3]. Metals oxides (e.g. titania, zinc oxide...) were also added to the virgin resin to improve mechanical and antibacterial properties [4]. Other examples are represented by natural fillers such as wood flour, cotton and flax fibers and other agricultural waste [5–7]. Moreover, other studies focused on the use of recovered plastic waste and their reuse in 3D printing [8,9]. These studies showed how the use of filler can bring both structural and environmental benefits. Despite the strong interest, literature still lacks of studies concerning effective ways to reuse CFRP manufacturing waste and ways to further improve SLA production process sustainability.

The present study aims at addressing these issues related to 3D printing and composite materials waste management by investigating the feasibility of using FRP scraps as fillers in photopolymer resin used in Masked Stereolithography (M-SLA) 3D printing. To the authors knowledge, this work represents a novelty in scientific literature, as it investigates the direct reuse of FRP machining scraps in a 3D printing process, combining experimental mechanical characterization with a cradle-to-grave environmental assessment.

Composite waste in the form of powder obtained from drilling operations was added at different weight percentages to neat photopolymer resin. Hence, mechanical characterization tests and sustainability analysis were carried out to determine whether this approach can provide a sustainable alternative to traditional 3D printing materials.

2. Materials and Methods

2.1. Materials and 3D printing technology

The main material used in the present study is the Elegoo translucent standard commercial resin. It is a LCD photocurable polymer that can be used in Masked stereolithography processes, and it mainly consists of epoxy acrylate resin and monomers and 3-5% by weight of photoinitiators, required to trigger the polymerization process. A translucent resin was selected to enhance contrast with the filler to better analyze its dispersion and interfacial interaction.

In order to improve the sustainability of the raw materials and reduce the consumption of neat resin, the resin was filled with fiber-reinforced scraps obtained from chip-removal manufacturing processes. Specifically, epoxy resin composites reinforced with basalt fiber were used. Tubular components were produced via a filament winding process starting from composite towpreg with a fiber and resin mass fraction equal to 72% and 28%, respectively. After the winding and oven curing process, the components were subjected to a drilling process as in common industrial practices for assembly or installations purposes. Figure 1 shows the drilling process of the FRP tubular component. This results in waste material in the form of powder and short fibers that was employed as a filler in the present study.

Starting from the resin and the filler, several material configurations were prepared to evaluate the effect of varying filler content within the resin on the mechanical properties of the resulting components. More specifically, 0% (neat resin), 0.5%, 1%, 2%, 2.5% and 5% filler content in weight were added to the resin. For each configuration, raw resin and FRP powder were weighed to obtain the target filler/resin weight and mixed together by means of mechanical stirrer. Hence, the so obtained composite material was poured within the tank of a Stereolithography (SLA) machine for the specimens production. Hence, 1 hour was waited before starting the printing process to allow the resin to set and let possible gas bubbles escape.

The Elegoo Saturn 4 Ultra LCD 3D printer was used for the present study. The machine features a Masked Stereolithography (M-SLA) that exploits a UV LED lights produced via a LCD screen to cure the photopolymer and produce the component layer by layer. The light is used to selectively harden a layer of the polymer according to the information provided by a digital instruction file, in turn based on a STL model of components to be produced. As each layer is completed, the building platform rises slightly to allow liquid material to get in contact with the LCD and to harden the next layer. The process continues until the final component is completed. 5 specimens at the time (either flexural or tensile specimens) were produced via M-SLA. Layer height equal to 0.05 mm, bottom exposure of 30 s, normal exposure of 8 s were set as process parameters and were kept the same for all the material configurations.

After the printing process, two postprocessing steps were carried out. At first, the produced parts were washed in isopropyl alcohol for 5 minutes to remove excess uncured resin. After that, the parts were subjected to UV lights for 30 minutes to ensure complete curing of the resin. The Elegoo X bundle, consisting of a washing and curing machine, was used for the postprocessing.

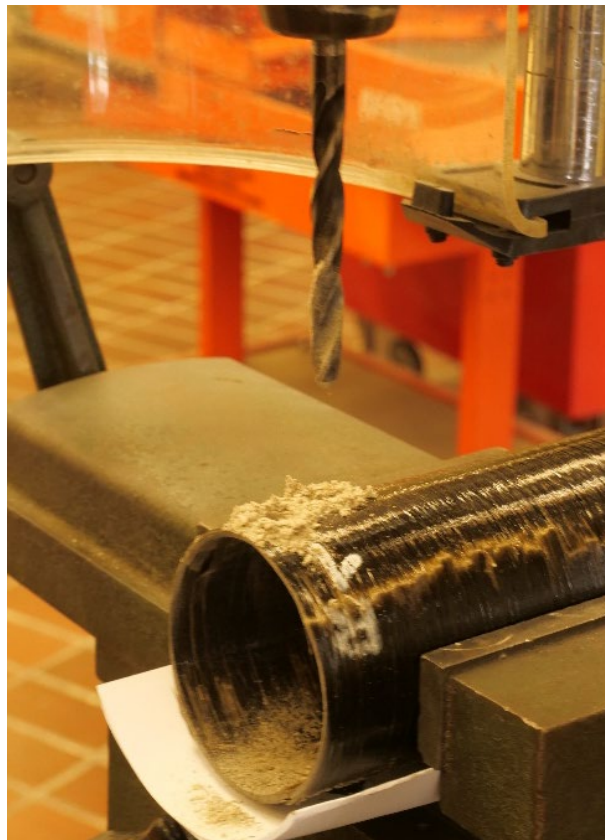


Fig. 1. Drilling process and scraps production for the tubular FRP composites.

2.2. Mechanical characterization

Tensile and flexural tests were carried out to evaluate the mechanical properties of the resin with different filler content. Neat resin samples were also produced to evaluate the raw materials properties and provide a baseline for the study. This was used as a reference to evaluate the effect of the FRP filler addition.

Type IV dog bone specimens were produced according to the ASTM D638-22 standard for tensile properties of plastics. The specimens have thickness, gauge length, and width equal to 3.2 mm, 33 mm and 6 mm, respectively. 5 specimens for each material configuration were printed and tested using a MTS810 universal testing machine. A crosshead speed equal to 5 mm/min was used for the test in line with the standard recommendation. An extensometer and a load cell were used to register data and create stress-strain curves for each specimen. Hence, a spreadsheet was used to evaluate the most relevant mechanical characteristics such as Ultimate Tensile Strength (UTS), Elastic modulus (E) and maximum strain.

Figure 2 presents the tensile specimens right after the printing process on the building platform (Figure 2a) and a detail of the dog bone specimens with recovered composite filler (Figure 2b). Similarly, the ASTM D790-17 standard was followed to assess the materials flexural properties. As for the tensile tests, 5 specimens for each condition were produced and tested with a speed equal to 2 mm/min. Specimens with thickness, width and length equal to 3.2 mm, 13 mm and 127 mm respectively were produced.

Fracture surfaces of the specimens were observed by means of the Leica M205 C Stereomicroscope to investigate fracture mechanisms and interaction between the filler and the matrix resin.

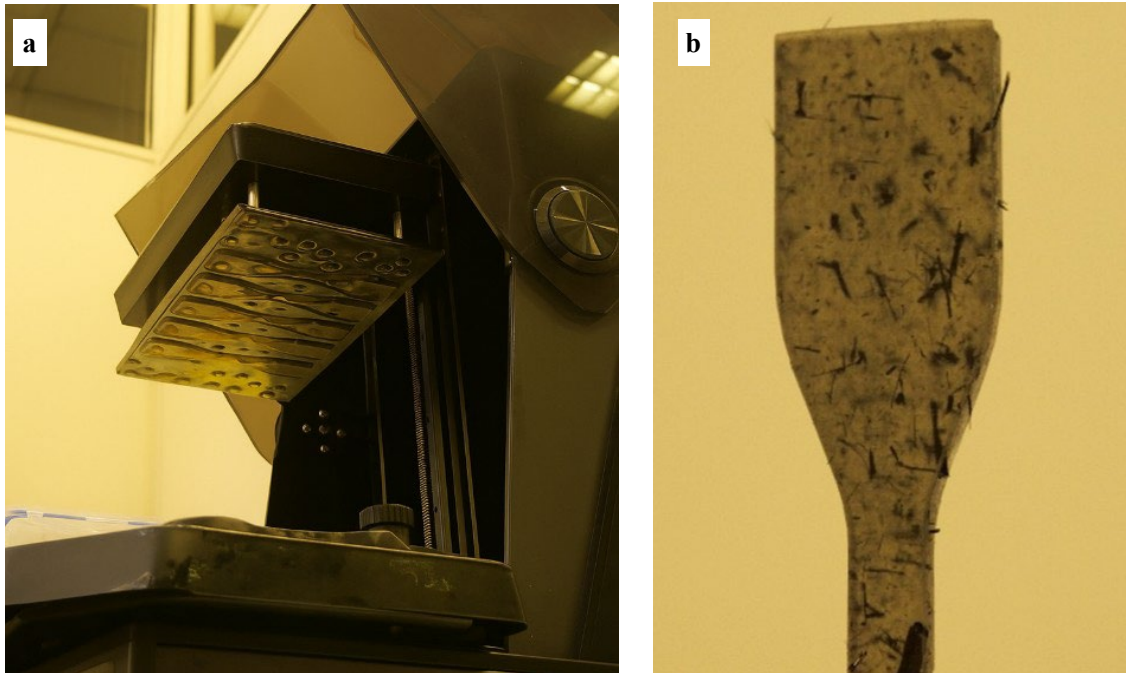


Fig. 2. 3D printing of tensile specimens (a) and detail of a printed specimen.

2.3. Life Cycle Assessment analysis

The environmental sustainability of the 3D printing process and the reuse of composite waste as filler was evaluated by means of the Life Cycle Assessment (LCA) methodology. The framework proposed by the ISO 14040-14044 standards was followed, and the four iterative phases were carried out: Goal and Scope Definition, Life Cycle Inventory, Life Cycle Impact Assessment (LCIA), and Results Discussion. These phases are detailed in the following paragraphs.

Goal and scope Definition

The present LCA analysis aims at evaluating and comparing the environmental impacts associated with the production of components produced via M-SLA using raw materials constituted by variable percentages of raw resin and recovered filler material. In particular, the analysis aims at understanding the environmental implications of using recovered FRP waste obtained from machining processes in 3D printing processes.

To do so, the Functional Unit (FU) is defined as the production of a tensile specimen with dimension defined by the ASTM D638 standard by means of a M-SLA process. Initially, the FU only considers geometric requirements, and it is suitable to compare components that do not have strict structural requirements (e.g. aesthetic components or prototypes).

A “Cradle to Grave” approach was followed and the following phases were included within the study: raw materials (resin and filler) production and transport, packaging materials, 3D printing phase, post processing (including washing and post curing) and End of Life (EoL). No impacts associated with the use phase of the components were considered as they would have been negligible for selected FU; this also contributes to give generalizability to the results as specific use phase impacts would strongly change depending on the application of the produced components.

In addition to the initial investigation, a subsequent LCA analysis was conducted to assess the environmental performance of components characterized by different filler content with defined structural requirements. Hence, an additional LCA and FU were defined as a tensile specimen that exhibits a strain equal to 0.6% when subjected to a load of 363 N. These values were based on the tensile test results of the neat resin specimens and were considered a reference for all scenarios.

Since the materials with different filler weight percentage have different mechanical properties (as reported in the results section), the FU should have different thickness depending on the material considered. The cross-section area of the different specimens was hence calculated considering simple mechanical relationship (i.e. Hooke's law, stress definition) and the elastic moduli of the different materials. This results in parts with bigger cross sections as the elastic modulus decreases. Since the neat resin scenario was selected as reference, the cross section of the part in this scenario is the same considered for the previous LCA (i.e. for nonstructural components).

This approach allowed to compare components with the same stiffness across different scenarios. In this way, it was possible to evaluate the environmental behavior of the FRP scraps reuse under different functional conditions (i.e. for aesthetic components or structural ones) [10]. The same scenarios previously described were considered in both analyses.

Life Cycle Inventory

Laboratory measurements were carried out to gather primary data related to the different produced specimens. The dimensions and weight of parts with different filler weight content were measured using a caliber and precision scale. Hence, the weight of each constituent (resin and filler) was calculated considering their nominal weight in percentage. The Ecoinvent 3.8 database was employed to model the resin production. For what concern the FRP filler, a cut off approach was considered and no burden related to its production were considered. This material is today typically disposed of in landfill or incineration facilities, so no impacts were considered associated with its production. Similarly, resin packaging (HDPE bottle, cardboard box, plastic wrap) was weighed and modelled according to the commercial database. The impacts of packaging were allocated to the functional unit considering the size of the resin bottle (0.5 l) and the weight of resin used for each material configuration.

Transport impacts were evaluated considering the material supplier, their distance with the production facility (considered in Center Italy) and Ecoinvent datasets related to road and air transport.

Energy consumption for the printing and post processing phase was directly measured using a power meter; these phases are considered the same for all the scenarios. Hence, the Ecoinvent dataset for electric energy (low voltage energy mix, Italy) was used. Isopropanol use was evaluated considering the washing tank capacity and the number of washing cycles possible before solvent substitution is required.

For all materials configurations EoL was modelled as landfill disposal modelled according to the Ecoinvent database.

Table 1 summarizes the main LCI data.

Life Cycle Impact Assessment

The LCA dedicated software SimaPro was employed to model the scenarios with different composite materials. The software is equipped by default with the Ecoinvent database.

The Global Warming Potential impact category, calculated according to the IPCC (International panel on Climate Change) methodology with a 100 years time horizon was considered for the analysis. This is one of the most commonly used impact categories in literature for LCA analysis concerning composite materials and 3D printing.

After the software modelling phase, the results were exported to a spreadsheet for further analysis and graphical representation.

Table 1. Main LCI data.

LIFE CYCLE INVENTORY				
Filler weight in percentage	Part weight (g)	Filler Weight (g)	Resin weight (g)	
0.0%	6.082	0.000	6.082	
0.5%	6.034	0.030	6.064	
1.0%	6.025	0.061	6.045	
2.0%	6.010	0.123	6.008	
2.5%	6.072	0.154	5.989	
5.0%	6.021	0.310	5.895	
Energy consumption (kWh)		Resin transport (km)		
Printing machine		13000 plane		
Washing		430 truck		
Curing		0.018		
Packaging (g) for SLA resin (0.5 kg)				
primary packaging (HDPE bottle)		82		
secondary packaging		58.56		
Plastic wrap		7.1		

Results and Discussion

Mechanical characterization

Figure 3 presents the results of the tensile tests for the materials characterized by increasing recovered filler weight content. Figure 3a reports typical stress-strain curves for the different specimens. It can be noted that the neat resin (0%) exhibits the highest tensile strength and strain at break amongst the presented curves; in line with the typical behavior of polymeric resins, the curve exhibits an initial linear elastic region, followed by a plastic deformation stage and finally failure.

The use of the recovered filler substantially reduces the UTS and strain at break of the resin.

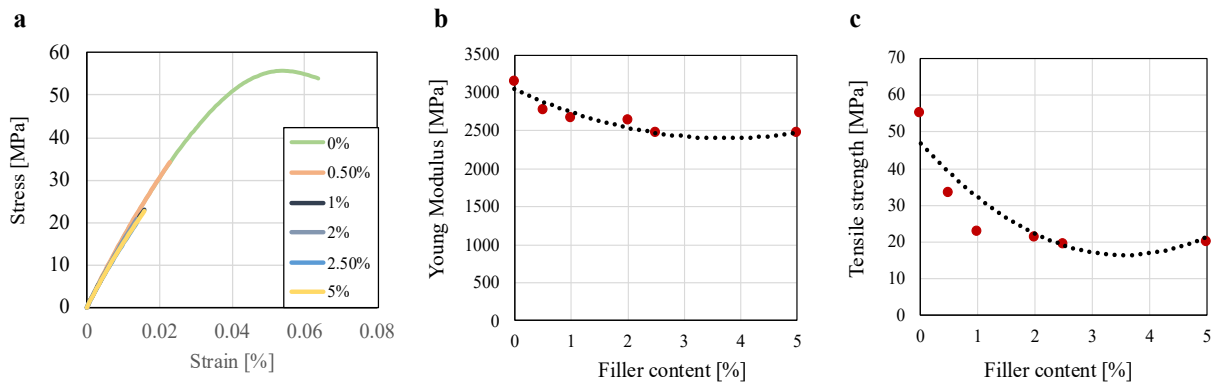


Fig. 3. Results of the tensile test for the specimens with increasing recovered filler content: a) typical stress-strain curves for the different materials; b) Elastic modulus as a function of the filler content; c) Tensile strength as a function of the filler content.

The initial slope of the curve (initial elastic region) is similar to the different alternatives. This can be also seen in Figure 3b, in which the elastic modulus is plotted as a function of the filler weight content. The Elastic modulus is reduced as the filler content increases, ranging from 3.16 GPa (Neat resin) to 2.47 GPa (5%), showing a maximum reduction equal to 21%. This decrease is more pronounced at low filler contents, with a reduction of approximately 16% between 0% and 1% filler, while at higher filler loadings (2.5–5%) the modulus values tend to stabilize, showing negligible further variation. On the other hand, the UTS strongly decreases with increasing recovered filler percentage. A reduction equal to 61% in UTS is observed between the neat resin and the 5% filler percentage resin (i.e. from 54.95 Mpa to 21.42 MPa). As for the elastic modulus, the reduction in mechanical properties is mainly observed when moving from the neat resin to low filler contents, with a decrease of about 58% between the neat resin and the 1% filler content. Beyond this point, the values tend to stabilize, remaining nearly constant for filler contents between 2.5% and 5%. The reduction in UTS can also be associated with the loss of strain at break with increasing filler content; this indicates embrittlement of the printed parts and therefore an early formation of cracks and their propagation [11–13]. The loss of mechanical properties with increasing filler content suggests that the FRP waste does not act as a reinforcement or a stiffening agent, but rather it lowers the load bearing capacity of the polymer. This behavior is expected when rigid particulate/short fibre is added to a polymer and poor bonding with the matrix is obtained. This, along with the possible presence of voids or trapped air introduced during filler/resin mixing is the probable cause of loss in mechanical properties in the recovered composite parts.

The behavior is also confirmed by the microscope image analysis as reported in Figure 4. Figure 4a shows the fracture surface of the neat resin where no clear defects of inclusion can be seen. On the other hand, Figure 4b reports the fracture surface of a tensile specimen with 5% in weight of filler. In this case, several discontinuities can be observed, directly associated with the presence of the filler. Some fibers appear to be well embedded within the matrix, while numerous white spots, corresponding to residual matrix fragments from the recycled filler, appear poorly bonded to the photocured resin. These regions likely act as stress concentrators and weak interfaces, representing the main cause of the observed reduction in mechanical performance.

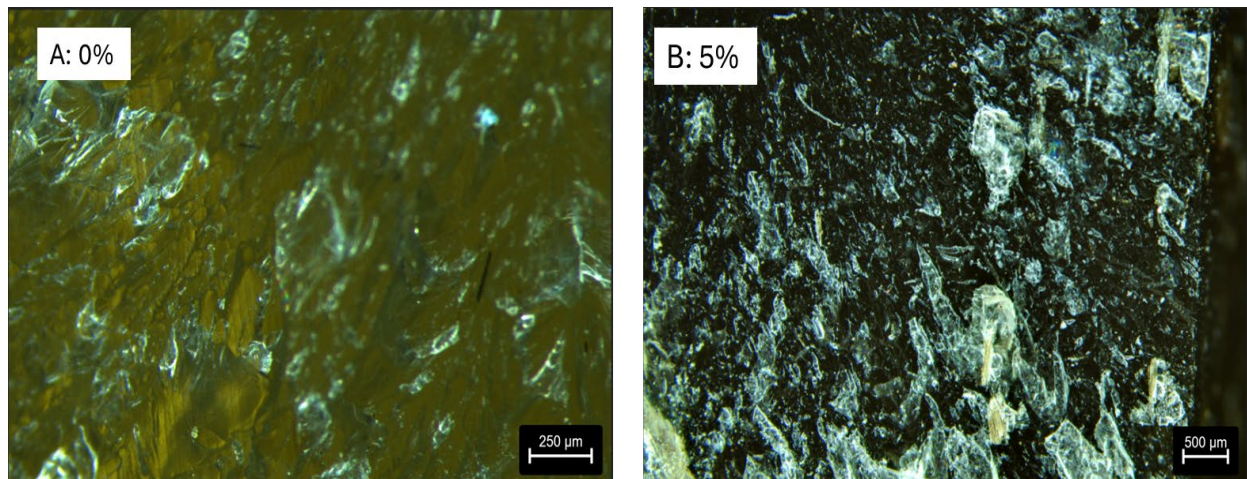


Fig. 4. Surface fracture of specimens with 0% (a) and 5% (b) of recovered FRP filler.

These regions likely act as stress concentrators and weak interfaces, representing the main cause of the observed reduction in mechanical performance.

Table 2 presents the results of the tensile and flexural tests in terms of max stress, strain, and elastic modulus. Overall, the same trend observed for the tensile properties can be seen for the flexural properties, with a decrease in load bearing capacity, ductility, and stiffness as the filler percentage increases. The max stress decreases by 63% from the neat resin to the 5% material configuration. On the other hand, the flexural modulus does not decrease significantly even for the highest filler percentage, with maximum reduction with respect to the neat resin equal to 5%.

Table 2. Tensile and flexural tests results.

Filler content	Tensile properties			Flexural properties		
	Max Stress [MPa]	Max Strain [mm/mm]	Elastic Modulus [MPa]	Max Stress [MPa]	Max Strain [mm/mm]	Elastic Modulus [MPa]
0%	54.95 (±3.70)	0.032 (±0.007)	3155.78 (±644.48)	94.71 (±6.02)	0.053 (±0.004)	2685.94 (±370.18)
0.5%	33.23 (±3.23)	0.014 (±0.004)	2775.10 (±172.97)	57.62 (±5.13)	0.023 (±0.005)	2846.78 (±129.67)
1%	22.86 (±2.42)	0.009 (±0.002)	2666.26 (±319.24)	67.50 (±3.66)	0.030 (±0.002)	2682.85 (±193.47)
2%	21.20 (±3.82)	0.008 (±0.001)	2640.82 (±237.15)	32.94 (±4.17)	0.014 (±0.002)	2584.40 (±167.51)
2.5%	19.50 (±3.55)	0.009 (±0.002)	2471.52 (±250.03)	44.49 (±6.00)	0.019 (±0.003)	2479.25 (±267.27)
5%	21.42 (±2.34)	0.009 (±0.001)	2471.00 (±213.57)	34.87 (±4.57)	0.020 (±0.002)	2540.78 (±233.23)

Sustainability assessment results

Figure 5 shows the results of the LCIA in terms of Global Warming Potential for the production of the tensile specimens constituted by resin with different filler percentage. The results show that the use of recovered composite material as a filler within the resin can provide environmental benefits. In fact, impacts reduction equal to 2% is obtained as the neat resin scenario is compared with the 5% filler one. Although the filler content is relatively low, this reduction is still significant, as the substitution directly affects the main contributors to the overall environmental impact, namely the production and transport of virgin resin. In fact, resin production and transport can account for up to 35% and 25% respectively of the total impacts of the scenarios. This is due to the synthetic origin of the resin (i.e. petroleum based) and the long transport distance required from the manufacturer (in China) and the final part production site. Hence, the reduction of the amount of virgin resin used in the process represents a good strategy to improve the sustainability of 3D printed components. On the other hand, the recovered filler has no impact, and its reuse also contributes to reducing the amount of material sent to landfill or incineration facilities. The printing and post-processing phases remain unchanged irrespective of the filler content in the resin, contributing approximately 15% and 20% of the total environmental impacts, respectively. These phases impacts are mainly related to the electric energy use of the machines (printers, washing and curing stations) and can be reduced if more sustainable energy sources (e.g. based on renewable energy) are used.

Although the environmental sustainability benefits increase with higher filler content, some technological limitations were observed. Increasing the filler percentage beyond 5% results in a more viscous resin, which becomes increasingly difficult to print successfully. As a result, the printed parts become more fragile, often showing poor surface quality and incomplete polymerization. Preliminary tests with 10% filler showed these issues, as printing could not be successfully completed. Therefore, further improvements are needed to mitigate the mechanical property reduction from a structural standpoint and to enhance the processability of resin with higher filler contents.

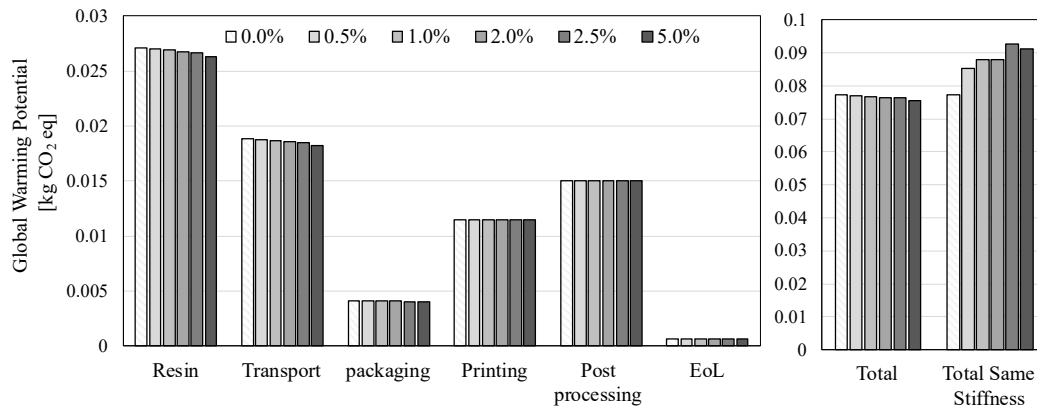


Fig. 5. LCIA results for parts constituted by different filler percentages with defined geometry or with defined structural requirements.

Regarding the structural components LCA analysis, the graph reports the total life-cycle impacts for the configurations ensuring the same stiffness (“Total Same Stiffness”).

Although this second LCA analysis for structural components involves a different functional unit, the total impacts of the structural components are displayed close together in the graph. This is to clearly show how impacts can vary depending on the structural requirements of the components used in different scenarios. This approach was necessary to provide a better understanding of the proposed recovery solution from both structural and environmental perspectives. This comparison is also possible since the two analyses consider the filler percentages.

A significant increase in environmental impacts is observed with the rise in filler content. This trend is explained by the reduction of the elastic modulus as the filler percentage increases, which requires larger cross-sections and therefore a higher resin consumption to achieve the same mechanical performance. Consequently, the impacts associated with resin production, material transportation, and the 3D printing process increase. Overall, an increase of approximately 18% in total impacts is observed when moving from the neat resin to the 5% filler configuration. However, the increase in impacts tends to stabilize beyond 2.5% filler, with even a slight reduction. This behavior can be attributed to the fact that the mechanical properties no longer decrease after that percentage, while the percentage of recycled filler increases, leading to a lower overall resin consumption.

Conclusions and Future Developments

This paper investigates the reuse of Basalt Fiber Reinforced Polymer powdered waste as a filler in a photocurable resin for M-SLA 3D printing processes. Resin with increasing filler content (0, 0.5, 1, 2.5 and 5 wt%) was used to produce tensile and flexural specimens to perform a mechanical characterization of the materials. In addition, a “from cradle to grave“ LCA analysis was performed to assess the environmental implications associated with the composite waste reuse.

The main outcome can be summarized as follows:

- The ultimate tensile strength strongly decreases very as small amounts of filler is added to the resin: a 58% reduction between neat resin and 1 wt% filler (from 54.95 MPa to 22.86 MPa), and overall, a 61% reduction at 5 wt% is observed.
- The elastic modulus decreases from 3.16 GPa (neat resin) to 2.47 GPa (5 wt% filler) (21% total reduction). The largest loss is observed between 0% and 1 wt% filler (16% decrease), while Elastic modulus values stabilize between 2.5–5 wt%
- If nonstructural components are considered, the use of the recovered filler provides a reduction in environmental impacts. Global Warming Potential is reduced by 2% with respect to the neat resin scenario as a 5% filler material is used. Despite the low filler content, benefits are registered as the use of filler reduces the impacts of the main life cycle contributors, namely resin production and transport.

- As structural components are considered, the use of recycled filler increases the GWP by up to 18%; this is due to the reduced stiffness of the materials with increasing filler content.

Overall, the reuse of FRP waste can provide environmental benefits, but some aspects of the process should be further investigated. Future work will focus on ways to improve the interface between the recovered material and the photopolimeric resin (e.g. via sieving of the waste powder) in order to improve the mechanical properties of the recovered materials. In addition, solutions to increase the filler content without reducing the processability of the resin will be investigated. These solutions, along with a continuous monitoring of the environmental impacts of the reuse process, can provide great sustainability benefits in the composites and 3D printing sectors.

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