



UNIVERSITÀ POLITECNICA DELLE MARCHE
Repository ISTITUZIONALE

Process parameters effect on environmental sustainability of composites FFF technology

This is the peer reviewed version of the following article:

Original

Process parameters effect on environmental sustainability of composites FFF technology / Bianchi, Iacopo; Forcellese, Archimede; Mancia, Tommaso; Simoncini, Michela; Vita, Alessio. - In: MATERIALS AND MANUFACTURING PROCESSES. - ISSN 1042-6914. - 37:5(2022), pp. 591-601. [10.1080/10426914.2022.2049300]

Availability:

This version is available at: 11566/297240 since: 2024-12-09T14:58:24Z

Publisher:

Published

DOI:10.1080/10426914.2022.2049300

Terms of use:

The terms and conditions for the reuse of this version of the manuscript are specified in the publishing policy. The use of copyrighted works requires the consent of the rights' holder (author or publisher). Works made available under a Creative Commons license or a Publisher's custom-made license can be used according to the terms and conditions contained therein. See editor's website for further information and terms and conditions.

This item was downloaded from IRIS Università Politecnica delle Marche (<https://iris.univpm.it>). When citing, please refer to the published version.

(Article begins on next page)

Process parameters effect on environmental sustainability of composites FFF technology

Abstract

The present study aims at investigating the effect of process parameters on environmental sustainability of 3D printing of short fiber-reinforced polymer composites. A preliminary study allowed defining the printing parameters, in terms of printing speed, extrusion temperature and layer thickness, which guarantee low energy consumption. Then, the environmental behaviour of two 3D printable composite materials, glass fiber-reinforced (GlassPA) and carbon fiber-reinforced (CarbonPA) polyamide, was investigated using the Life Cycle Assessment methodology, to provide a comprehensive overview of the considered materials and to support the sustainable development of industrial additive manufacturing processes. The functional unit was chosen taking into account the mechanical properties of the two short fiber-reinforced composites. To this purpose, tensile and flexural tests were performed on specimens produced by Fused Filament Fabrication process to evaluate the mechanical properties of printed materials. Scanning electron microscopy was used to observe the different filament morphology affecting the materials performances. Experimental tests showed that CarbonPA exhibits mechanical performances higher than those of GlassPA. Due to the weight reduction that can be accomplished by means of carbon fibers, CarbonPA results the most environmentally friendly alternative in tensile loads applications. On the contrary, for flexural loads applications, GlassPA exhibits lower environmental footprint.

Keywords

GFRP, CFRP, 3D printing, Parameters, LCA, Sustainability, Tensile, Flexural, SEM.

Introduction

In recent years, the automotive and transport industries are committed to facing new challenges deriving from the international regulations that force the development of energy-efficiency strategies to reduce emissions. The vehicle weight saving, combined with the guarantee of high mechanical performances, is a key factor on which many engineers and researchers are dedicated in order to make transport safe

and energy efficient. ^[1] The vehicle manufacturers attention is particularly focused on the fiber-reinforced polymer composites (FRPC) since they can be used as effective materials for replacing metals in many applications in which lightness and high strength are required.^[2-4] Among FRPC materials, carbon fiber-reinforced polymers (CFRP) and glass fiber-reinforced polymers (GFRP) can strongly decrease car weight.^[5,6] Several traditional processes, such as resin transfer molding, hand layup method, automated fiber placement, etc. were used to manufacture components in FRPC materials.^[7,8] Furthermore, the three-dimensional (3D) printing is recently used to additively manufacture parts in fiber-reinforced composite materials.^[9-13] The growing interest in environmental issues must push researchers to consider products sustainability at the same time as mechanical performance.^[14] Indeed, carbon fibers are typically derived from fossil resources and their transformation is based on energy intensive processes conducted at very high temperatures (above 2000°C). Differently, glass fibers production is a less impactful process owing to the availability of raw materials (principally SiO₂, Al₂O₃ and CaO) and to the easier transforming process based on an extrusion at temperatures below 1500°C. In this context, Life Cycle Assessment (LCA) is a well-known technique used to conduct a comprehensive evaluation of the environmental impacts of products and processes.^[15] In the scientific literature, no research that correlates the mechanical properties and environmental impacts of 3D printed components in glass and carbon fiber-reinforced polymers are available. Indeed, even though carbon fiber composites are more performant than glass fiber ones, their elevated impacts make the material-choosing process extremely complicated. Therefore, the choice of the most environmentally friendly fiber-reinforced composite material for a defined application is not obvious for designers. The environmental sustainability of a manufacturing process is also significantly influenced by the process parameters used.^[16] No studies concerning the analysis of the effect of these printing parameters on the energy consumption were found. This is a key aspect to develop more sustainable 3D printing processes providing additional information with respect to the discussion of the effects of manufacturing parameters on mechanical properties.

In this framework, the present work aims at investigating the effect of process parameters on the environmental sustainability of 3D printing of two short fiber-reinforced composites. A preliminary

investigation was carried out in order to define the printing parameters, in terms of printing speed, extrusion temperature and layer thickness, which guarantee low energy consumption. Then, the mechanical properties and environmental impacts of 3D printable GlassPA and CarbonPA composites materials were investigated. To this purpose, specimens were produced using a Fused Filament Fabrication (FFF) method. Tensile and flexural tests were performed to evaluate the mechanical performances of the printed materials. The environmental behaviour of both CFRP and GFRP was investigated using the LCA methodology; a cradle to grave method was chosen to evaluate all the relevant input of the parts life cycle, from raw materials extraction to products disposal. Both tensile and flexural specimens were analyzed from environmental point of view. Inventory data were collected from literature research, commercial databases and from direct measurements of the production phases.

Materials and methods

CFRP and GFRP composites investigated in the present study are the commercial CarbonPA and GlassPA. They consist in a polyamide (PA) matrix reinforced with 20% in weight of short carbon fibers and with 30% in weight of short glass fibers, respectively. The fiber volume fractions of CarbonPA and GlassPA are equal to 14% and 16%, respectively. Such composites, due to their low weight and high strength, are mainly used as metal replacement in automotive, military and aerospace applications.

Filaments in CarbonPA and GlassPA, supplied in form of spools, were printed using the Roboze One+400 3D printer based on the FFF additive manufacturing technology. Since the polyamide is strongly subjected to humidity absorption, which causes a significant decrease in mechanical properties, both composite materials were dried for about 2h at the temperature of 120°C before printing; moreover, during the printing phase, CarbonPA and GlassPA filament spools were kept at a constant temperature of 70°C inside a dryer, external to the 3D-printer, to avoid humidity adsorption.^[17] Filament extrusion of both fiber-reinforced composite materials was performed at a temperature higher than the glass transition temperature (T_g) of polyamide. The process parameters used to print both CarbonPA and GlassPA were selected after carrying out a preliminary investigation, based on the evaluation of the energy consumption of the additive manufacturing operation as a function of process parameters. Specifically, different printing speeds, temperature, and layer thickness values were taken into account.

Table 1 summarizes the additive manufacturing process parameters imposed. Samples were 3D printed on a Bakelite bed. A raft of 8 mm and a lower support of 3 mm were provided to avoid the warping effects typical of polyamide, which tends to deform during shrinkage, and to enhance the adherence of the parts to the printing bed.

Table 1: Additive manufacturing process parameters used to print CarbonPA and GlassPA.

The mechanical properties of 3D printed CarbonPA and GlassPA composites were investigated by exploiting both uniaxial tensile and flexural tests, carried out on 3D printed samples in FRPC in accordance with the ASTM D3039 and ASTM D790-17 international standards, respectively, using the servo-hydraulic MTS 810 machine. Samples were obtained in the flat orientation, coincident with print bed. The gauge length, width and thickness of tensile samples were equal to 80 mm, 10 mm and 4.5 mm, respectively. The grip section was increased both in thickness (6 mm) and width (20 mm) to prevent failure due to the high clamping force applied by the gripping mechanism. As far as the flexural tests are concerned, a three-point loading configuration, characterized by a loading nose and two supports with radii of 5.0 ± 0.1 mm, with cylindrical and finely ground contact surfaces, was used; the support span was equal to 68 mm. Samples were characterized by a length, width and thickness of 80, 12.7 and 4 mm, respectively. For both tensile and flexural tests, the crosshead speed was kept constant and equal to 2 mm/min. Three repetitions per test condition were executed. The cross- and longitudinal sections of the CarbonPA and GlassPA filaments before 3D printing process were observed using ZEISS and VEGA scanning electron microscopes. Samples were coated exploiting a metallization process to make them conductive and identifiable by the scanning electron microscopy (SEM).

In order to investigate the environmental behaviour of both CarbonPA and GlassPA, the impacts analysis was conducted following the standardized framework of LCA described by the ISO 14040 and 14044. LCA is a well-known methodology developed to assess the environmental effects of a product, a process or a service during their life span. The main steps of the LCA methodology were followed:

- 1) Goal and scope definition: the objective of the study, the product system (or multiple systems in case of comparative analysis), the system boundaries and the functional unit (FU) are defined.

- 2) Life Cycle Inventory: it consists in the data collection regarding all inputs (raw materials, energy consumptions, ...) and outputs (waste, emissions, ...) of the system boundaries.
- 3) Life Cycle Impact Assessment (LCIA): data gathered during the inventory phase are converted into potential environmental impacts according to different impacts categories.
- 4) Interpretation of results: the LCIA results are interpreted and completeness, sensitivity and consistency checks are made to improve the analysis reliability. Lastly, conclusions, limitations and recommendations are proposed considering the goal and application of the study.

Results and discussion

FFF parameter effects on environmental sustainability

A preliminary investigation, based on the effect of the printing parameters on energy consumption, was performed to evaluate the process parameters for the 3D printing of GlassPA and CarbonPA filaments. Different printing speed, layer thickness and extrusion temperature values were imposed and the energy consumption was measured. Figure 1a shows the printing times required to obtain a tension sample as a function of printing speed and layer thickness. For a given printing speed, the printing time values reduce as the layer thickness increases, since a higher quantity of material is deposited in each layer, allowing the achievement of the desired specimen height with a reduced number of layers. Furthermore, for a given layer thickness value, the printing time decreases with rising printing speed. At the highest printing speed investigated, the specimens obtained with layer height equal to 0.18 and 0.24 mm showed a poor surface finish. Such result can be attributed to the rise in surface roughness values with increasing layer thickness, as shown by Kandananond^[18] and Chaidas at al.^[19] Since surface roughness strongly affects both the interaction between the part and environment and the crack formation, these conditions were not considered in such study (values marked with X in the Figure 1a). For each 3D printing condition, the electric energy consumption values were directly measured. Such results were plotted as a function of layer thickness at different printing speeds (Figure 1b). The lowest analysed printing speed (30 mm/s) corresponds to a much higher electrical energy consumption with respect to all the other alternatives. In fact, keeping the layer thickness constant, the energy consumption decreases as the

printing speed increases. Therefore, excluding the printing speed of 90 mm/s, which generates a poor surface finish, setting a high printing speed of 70 mm/s leads to both high productivity and low electric energy consumption. As far as the layer thickness is concerned, Figure 1b also shows that, for a given printing speed, low electric energy consumption can be obtained with high layer thickness values. Unfortunately, as shown in Figure 2, high layer thickness produces a high number of canyons between adjacent filaments. On the contrary, when the lowest layer thickness is set, a more compact material deposition occurs covering a small area and, consequently, very small internal gaps between adjacent filaments generate. Then, the cross-section area increases, leading to improved mechanical properties. To reduce the voids formation in the 3D printed material, which could also worsen the mechanical properties and surface finish of the component ^[20], it is advisable to avoid setting the highest layer thickness investigated (equal to 0.24 mm), but prefer a layer thickness of 0.18 mm. Furthermore, this choice does not entail significant increases in electric energy consumption for the highest printing speeds considered in the present work (70 and 90 mm/s).

Printing jobs were carried out varying the extrusion temperature from 230°C to 300°C, measuring the electrical energy consumption. Figure 1c shows the energy consumption as a function of the extrusion temperature. It can be observed that, keeping other parameters fixed, energy consumption increases as the extruder temperature increases. However, temperature does not have a remarkable influence on the printing phase energy consumption; in fact, the recorded energy consumption increment is only of 7.4% over a temperature variation of 70°C. For the lowest tested temperature, insufficient adhesion between the building plate and the raft caused shifts of the samples, compromising the success of the printing process. At 260 °C, the printing process is properly executed and specimens with good surface finish are obtained. Beyond this temperature, there is no noticeable improvement in the quality of the manufactured parts. For what concerns the building plate, the temperature has been set at 80°C; decreasing this temperature leads to warpage phenomena which may determine the non-conformity of the parts to the project requirements.

Figure 1: Effect of: (a) printing speed on printing time, and (b) layer thickness and (c) extrusion temperature on electric energy consumption

The investigation on the printing parameters, therefore, led to the choice of the printing speed equal to 70 mm/s, the extrusion temperature of 260°C, and layer thickness of 0.18 mm that allow to obtain a compromise between aesthetic appearance, mechanical properties and energy efficiency. Regarding printing infill arrangement, a complete infill of 100%, with a linear pattern, was set in order to minimize the number of micro-voids and align the filaments and the reinforcement fibers along the longitudinal axis of the samples, improving their mechanical properties in that direction. Since the two composite materials are characterized by the same polyamide matrix, specimens were printed with the same optimal printing parameters.

Figure 2: (a) Schematic representation of printed specimens and (b) effect of layer thickness on the canyon formation between adjacent filaments.

Figure 3 shows SEM images of the GlassPA and CarbonPA filaments before the 3D printing operations. The original filaments, characterized by short fibers randomly arranged, exhibit a porosity spread over the cross section of the filament (Figures 3a and 3c). Furthermore, it can be observed the high number of fibers dispersed in the polymeric matrix and how they are mostly aligned in the longitudinal direction as a result of the extrusion process used to produce the filaments (Figures 3b and 3d). A huge quantity of micro-voids, due to the fiber pull-out, can be seen in both filaments (Figures 3a and 3c); a higher number of macro-voids can be observed in the CarbonPA filament. However, these voids are sensibly reduced during the 3D printing due to the decrease of filaments diameter from 1.75 mm to 0.4 mm.

As far as the mechanical properties of CarbonPA and GlassPA composites is concerned, uniaxial tensile tests and flexural tests were carried out on 3D printed specimens. In accordance with the results obtained from the study on the effect of process parameters, the printing speed and layer thickness were set equal to 70 mm/s and 0.18 mm, respectively.

Figure 3: SEM images of filaments before 3D printing: (a) cross section and (b) longitudinal section of GlassPA; (c) cross section and (d) longitudinal section of CarbonPA.

Figure 4: Comparison between typical stress vs. strain curves of PA, CarbonPA and GlassPA: (a) tensile and (b) flexural behaviour (printing speed: 70 mm/s; layer thickness: 0.18 mm).

Figure 4a shows the typical nominal stress vs. nominal strain curves, provided by performing tensile tests on GFRP and CFRP 3D printed samples. The stress - strain curve obtained by testing the unreinforced polymeric material is also shown in Figure 4a, as a term of comparison. Irrespective of the FRPC material investigated, it can be observed that the curves of CarbonPA and GlassPA exhibit a behavior similar to the one shown by short fiber-reinforced polyamide. Both composite materials are characterized by a brittle behaviour, with low ultimate elongation values of about 2.1 and 2.6%, respectively; on the contrary, the unreinforced polyamide exhibits a ductile behavior. Both reinforced composites are characterized by a marked increase in both ultimate tensile strength (UTS) and elastic modulus (E) as compared to the un-reinforced polyamide ($UTS_{PA} = 44.9$ MPa and $E_{PA} = 0.49$ GPa).^[21] In detail, the increase in UTS and E obtained using carbon fibers ($UTS_{CarbonPA} = 124.0 \pm 4.7$ MPa and $E_{CarbonPA} = 11.6 \pm 0.4$ GPa) is more marked than that obtained by reinforcing matrix with glass fibers ($UTS_{GlassPA} = 67.7 \pm 3.4$ MPa and $E_{GlassPA} = 5.5 \pm 0.3$ GPa).

As far as the flexural performances of 3D printed filaments are concerned, CarbonPA and GlassPA exhibit similar behaviour (Figure 4b).^[22] As a matter of fact, both composites are characterized by an increase in stress with strain until a maximum value (ultimate flexural strength, UFS) is reached; the values of the ultimate flexural strength and elastic modulus obtained using carbon fibers ($UFS_{CarbonPA} = 136.0 \pm 7.3$ MPa and $FE_{CarbonPA} = 6.6 \pm 0.52$ GPa) are slightly higher than those obtained using glass fibers ($UFS_{GlassPA} = 119.8 \pm 6.7$ MPa and $E_{GlassPA} = 4.1 \pm 0.5$ GPa) indicating that the flexural behavior of fiber-reinforced polymer materials is more influenced by the matrix than by the fibers.

Environmental assessment of CFRP and GFRP

As far as the Life Cycle Assessment is concerned, in accordance with the standardized procedure, the first step of the analysis was to define the goal and scope. The objective of the analysis is to evaluate and compare the environmental behaviour of two composite materials: CarbonPA and GlassPA. The study aims to determine which of these two materials is the most environmentally friendly alternative for additive manufacturing applications. The FU was defined as “the production of a tensile specimen that exhibits a maximum strain equal to 2.55% when subjected to a tensile load of 3.1 kN and has a length of 170 mm”. Considering that the two materials have different densities and show different

mechanical properties, the FU has a different weight and sections area in the two scenarios. The weight of the functional unit in the different scenarios was calculated considering simple mechanical relations (i.e. Hooke's law, mass-density relation). Using GlassPA, the mass required to achieve the defined stiffness is 18.21 g while, for the CarbonPA, a mass equal of only 9.21 g is needed. Two different scenarios were evaluated to represent possible production processes: Scenario 1 considers the production of the specimens in GlassPA and Scenario 2 considers the production of the specimens in CarbonPA. Moreover, a sensitivity analysis, in which a new functional unit was defined on the basis of the flexural tests, was performed. The present LCA can be classified as "cradle to grave" analysis since it considers life cycle phases of the different specimens from the extraction of raw materials to their final disposal.^[23,24] Figure 5 shows the system boundaries for all the considered scenarios. Double colored blocks represent life stages that are included in both Scenarios 1 and 2.

Figure 5: System boundaries of all the considered scenarios.

The first phase of each process consists in the raw materials extraction and the production of the reinforced PA filament for the printing process. It considers the production of the fibers (1. glass fibers and 2. carbon fibers) and matrix as well as the filament extrusion process and materials transport. CFs were considered to be produced from Polyacrylonitrile (PAN) precursor; impacts related to PAN production, stabilization, carbonization and final treatments of the fibers were included in the analysis.^[25] Glass fiber production consists in melting raw materials such as SiO₂, Al₂O₃ and CaO in a furnace (i.e. a recuperative or oxy-fuel fired furnace). The molten glass is extruded in order to produce thin filaments of about 10 μm. A sizing agent is applied before gathering the fibers. Impacts related to the glass fibers were retrieved from the Ecoinvent database considering European glass manufacturing industry data. Before printing, both composite filaments were dried for 2 hours at 120°C to reduce their humidity content. This is a critical step because, as reported in previous literature studies, polyamide composites tend to absorb humidity from the air with a subsequent reduction of their mechanical properties.^[17] During printing, filament was kept in a heated atmosphere at 70°C to avoid water absorption (filament heating). The printing phase considers the energy consumption of the machine and the impacts related to the production and transport of the Bakelite build plate. The energy consumptions

of the printing and heating phases depend on printing time and, consequently, on the printed part weight. The useful life of the specimens was considered out of the system boundaries as it would lead to negligible impacts. The impacts related to the production of the machines used (i.e. printing machine and furnace) were neglected due to their long useful life with respect to the production time required for a single specimen.^[26] Landfill disposal was considered in both scenarios as it is the most common End of Life (EoL) option for composites waste.^[27]

As far as the Life Cycle Inventory is concerned, data from different sources were considered: primary data were directly measured during the production processes while secondary data were collected from literature and Ecoinvent 3.1 commercial database (Table 2). More specifically:

1. Data related to the input material of Scenario 1 were measured. Concerning Scenario 2, input materials were estimated considering that the FU must have the same stiffness in all the scenarios.
2. The modelling of the CF is based on the study of Forcellese et al.^[25]
3. Modelling of the filament composition was based on the material datasheets.
4. Electric energy consumptions were measured through the power analyzer PQ824 by HT instruments.
5. Transport distance was estimated considering the geographical location of various suppliers.
6. Data related to raw materials (i.e. Bakelite, glass fibers, polyamide...), electric energy generation, impacts of the transport and EoL phases were retrieved from the Ecoinvent database.

Table 2: Inventory data.

The software SimaPro 9.1.0.11 was used to model the two considered production processes and to translate inventory data into possible environmental impacts. The environmental footprint was assessed considering two impact categories: the Cumulative Energy Demand (CED, in MJ) and the Global Warming Potential (GWP, in kg CO₂ eq). Such impact assessment categories were widely used in scientific literature to evaluate the environmental behaviour of composite part production as well as additive manufacturing technologies.^[28-33]

Figure 6: Life Cycle Impact Assessment results in terms of (a) CED and (b) GWP.

Results of mechanical test were used to define the FU of the environmental impact analysis. Figure 6 shows the results of Life Cycle Impact Assessment of the two Scenarios in terms of Cumulative Energy Demand (CED) and Global Warming Potential (GWP). In both cases, Scenario 2, related to the CarbonPA composite, is characterized by the lowest environmental impact. Regarding CED (Figure 6a), a total energy consumption of 6.19 MJ is associated with the GlassPA specimen; this value is around 22% higher than the energy required for the CarbonPA specimen life cycle which is equal to 4.85 MJ. As shown in Figure 6b, Scenario 2 provides a strong reduction of the environmental burdens even considering the GWP indicator (with emissions values of 0.32 kg CO₂ eq for the GlassPA and 0.25 kg CO₂ eq for the CarbonPA). The footprint reduction associated with Scenario 2 is mainly determined by the heating and printing phases of the specimens. As emerged from measurements, the energy consumptions of these phases have a linear relationship with the required printing time and, therefore, with the parts weight. Thus, the CarbonPA specimen, which is about 50% lighter than the GlassPA one, results in a significant reduction of the energy consumption of these production phases. The strong influence on the total environmental impacts of the printing and heating phases makes the optimal process parameters setting crucial; as shown in the preliminary analysis, incorrect choices of process parameters, such as extrusion temperature, printing speed and layer thickness, may result in a noticeable increase of the printing phase time and energy consumption, with a subsequent increment in environmental impacts. In addition, inappropriate parameters choices can lead to the fabrication of parts not conform to the design specifications (i.e. warped parts or poor surface quality) that must be discarded. This leads to material and energy waste.

Despite the significant weight difference of the different specimens, the raw material production phases have shown similar environmental impacts for the two scenarios, with percentage variations lower than 10% (Figure 7). As far as the matrix is concerned, it can be seen that polyamide production represents the main contribution on the total impacts of the filaments, accounting for more than 80% of the final values of the material CED and GWP for Scenario 1 and about 50% for Scenario 2. This is a valid result considering the high percentage of PA on the composition of the raw materials (70% and 80%) and its impacts per kg produced (119 MJ and 8.28 kg CO₂ eq).

Figure 7: Environmental impact contributions for raw materials production: (a) CED and (b) GWP.

As far as the reinforcement is considered, carbon fibers used in Scenario 2 have a very high impact per kg produced (493.6 MJ and 27.8 kg CO₂ eq) so, despite the small amount required (1.84 g), they account for 43% of the impacts of the materials in Scenario 2. On the other hand, glass fibers used in Scenario 1 have little influence on the total impacts since their production has a CED and a GWP equal to 30.9 MJ and 2.45 kg CO₂ eq per kg produced. However, the strong weight reduction obtained using carbon fibers allows to reduce the contribution of polyamide and, consequently, makes the CarbonPA the best environmental choice. As far as the printing stage is concerned, the filament extrusion impacts depend on the weight of the produced part, so Scenario 1 is characterized by a higher contribution than Scenario 2; the extrusion, transport and End of Life phases provide a negligible contribution to the total environmental load. The drying phase is the same for the two scenarios as, to avoid moisture absorption, the whole filament roll is dried before printing, so the energy consumption of this phase is not directly related to the produced component weight.

Depending on the specific application, 3D printed components can be subjected to different kinds of loads, i.e. flexural loads. For this reason, a sensitivity analysis was conducted to compare the environmental behaviour of two specimens characterized by the same flexural performance. This allows a more complete view of the environmental impacts of the two composite materials. A new functional unit was defined as the production of a 3D printed sample that, during a flexural test, exhibits a maximum strain of 2% when subjected to a load of 556.8 N (P) and has a length (L) of 100 mm. Once again, the weight of the FU varies in the two scenarios and depends on the density and flexural modulus of the two materials. To guarantee the minimum flexural stiffness defined for the FU, the specimens section dimensions were calculated considering the equation (1) (ASTM D790-17 standard) and the flexural modulus measured during the flexural tests:

$$\sigma_f = \frac{3PL}{2bd^2} \quad (1)$$

Moreover, the ratio between the width and dept of the section dimensions (b and d) was set equal to 4. Table 3 presents the dimensions and the weights of the different samples. As shown in Figure 4, CFs

are characterized by a high increase in tensile stiffness as compared to the glass fibers, while the improvement is less relevant in terms of flexural modulus. For this reason, if samples with the same flexural behaviour are considered, the weight saving obtained by using CarbonPA is not very significant (about 11%).

Table 3: Dimensions and weight of the two considered samples.

The system boundaries and the production phases considered in this analysis are the same as the previous one. However, since the produced samples vary, some of the material and energy inputs of the system change too. Therefore, the inventory data were updated; most of them, such as transport distance, drying energy consumption, filaments composition and data from the Ecoinvent database, remain the same. Data which depend on the samples dimensions and weights (i.e. printing and heating energy consumption and input materials) have been updated considering the new functional unit. Inventory data sources are the same detailed in Table 2.

Figure 8: Environmental impacts in terms of (a) CED and (b) GWP for all the phases considered within the system boundaries.

Figure 8 shows the results of the Life Cycle Impact Assessment conducted considering the new functional unit. For both CED and GWP, Scenario 2 exhibits the highest environmental impacts, with values about 20% higher than those related to the GlassPA sample (7.39 MJ and 0.39 kg CO₂ eq respectively, compared with 6.19 MJ and 0.317 kg CO₂ eq of Scenario 1). Due to the lower weight of the CarbonPA sample, the filament heating and printing phases have reduced impacts in comparison with the GlassPA alternative (lower weight corresponds to shorter printing time and, consequently, electric energy saving). However, this is negligible if compared to the strong impacts associated with raw materials of Scenario 2, that account for 3.67 MJ and 0.21 kg CO₂ eq.

Figure 9 shows a detailed comparison between the contributions of the raw materials production phases. CFRP has higher impacts mainly due to the reinforcement phase. In fact, despite being in lower percentage, CFs lead to impacts that are 10 times higher than those of GFs, i.e. 1.6 MJ for the CF vs 0.15 MJ for the GF. In addition, the environmental footprint related to the matrix is also higher in the

CarbonPA scenario; as a matter of fact, even if the CF-reinforced sample has a lower weight, the polyamide percentage it contains is higher. Differently from the tensile specimen case, the weight reduction achieved by means of the CFs is not enough to compensate for the greater impacts of the raw materials for the flexural specimens. Thus, the environmental friendliness of one material over another depends on the specific application and the operating loads the components must withstand.

Figure 9: Environmental impact contributions for raw material production: (a) CED and (b) GWP.

Conclusions

In this study, the effect of process parameters on environmental sustainability of 3D printing of short fiber-reinforced composites was studied. A short carbon fiber-reinforced polyamide (CarbonPA) and a short glass fiber-reinforced polyamide (GlassPA) were 3D printed by exploiting the FFF process. A preliminary investigation allowed defining the printing speed, extrusion temperature and layer thickness to make the additive manufacturing process energy-efficient. Then, the environmental behaviour of GlassPA and CarbonPA was investigated using the Life Cycle Assessment methodology. Tensile and flexural tests were carried out to evaluate the mechanical behaviour of the 3D printed specimens in CarbonPA and GlassPA; the environmental analysis was conducted in accordance with the LCA methodologies, defining the functional unit based on the evaluated mechanical properties. In addition, scanning electron microscopy analysis was performed on filaments before printing. The main results are reported as follows:

- For a given printing speed, the electric energy consumption during 3D printing process decreases as the layer thickness increases; such behaviour is more marked at low printing speeds;
- At an imposed layer thickness, the electric energy consumption during printing process diminishes with increasing printing speed;
- The optimal extrusion temperature, for both the materials, is 260°C. At lower temperature, a weak adhesion between the building plate and the parts was registered. At higher temperature, energy consumption increases but no clear improvements in parts quality were obtained;
- The surface finish decreases with increasing printing speed and layer thickness;

- Both CarbonPA and GlassPA are stronger and stiffer than unreinforced polyamide;
- CarbonPA is characterized by a resistance and a stiffness 83.1% and 110.9%, respectively, higher than GlassPA;
- CarbonPA exhibits a flexural resistance and a flexural modulus 13.5% and 62.4%, respectively, higher than GlassPA;
- CarbonPA and GlassPA filaments are characterized by a huge quantity of void before 3D printing;
- From the environmental perspective, the choice of the most sustainable material strongly depends on the stresses acting on the fiber-reinforced composite material: CarbonPA is more eco-friendly than GlassPA when tensile loads act on 3D printed components; on the contrary, GlassPA is characterized by a better environmental performance than CarbonPA if 3D printed components must withstand flexural stresses.

Future work will be dedicated to the evaluation of other reinforcements based on natural fibers, such as flax and hemp ones, which can guarantee noteworthy mechanical properties and, at the meantime, significant reduction in the environmental impacts.

Acknowledgments

The Grant of Excellence Departments, MIUR-Italy (ARTICOLO 1, COMMI 314-337 LEGGE 232/2016) is gratefully acknowledged.

Reference

- [1] Sathiyamurthy, R.; Duraiselvam, M. Selective Laser Ablation of CFRP Composite to Enhance Adhesion Bonding. *Mater. Manuf. Process.* **2019**, *34* (11), 1296–1305. DOI:10.1080/10426914.2019.1644453.
- [2] Mosleh, N.; Rezaoust, A. M.; Dariushi, S. Determining Process-Window for Manufacturing of Continuous Carbon Fiber-Reinforced Composite Using 3D-Printing. *Mater. Manuf. Process.* **2020**, *36* (4), 409–418. DOI:10.1080/10426914.2020.1843664.
- [3] Silvestri, A. T.; Papa, I.; Rubino, F.; Squillace, A. On the Critical Technological Issues of

- CFF: Enhancing the Bearing Strength. *Mater. Manuf. Process.* **2021**, 1–14.
DOI:10.1080/10426914.2021.1954195.
- [4] Kechagias, J. D.; Ninikas, K.; Petousis, M.; Vidakis, N.; Vaxevanidis, N. An Investigation of Surface Quality Characteristics of 3D Printed PLA Plates Cut by CO₂ Laser Using Experimental Design. *Mater. Manuf. Process.* **2021**, *36* (13), 1544–1553.
DOI:10.1080/10426914.2021.1906892.
- [5] Das, S. *The Cost of Automotive Polymer Composites: A Review and Assessment of DOE's Lightweight Materials Composites Research*; ORNL/TM-2000/283; Oak Ridge National Lab. (ORNL): Oak Ridge, TN, 2001. DOI: 10.2172/777656.
- [6] Aleksendrić, D.; Bellini, C.; Carlone, P.; Ćirović, V.; Rubino, F.; Sorrentino, L. Neural-Fuzzy Optimization of Thick Composites Curing Process. *Mater. Manuf. Process.* **2018**, *34* (3), 262–273. DOI:10.1080/10426914.2018.1512116.
- [7] Forcellese, A.; Mancina, T.; Russo, A. C.; Simoncini, M.; Vita, A. Robotic Automated Fiber Placement of Carbon Fiber Towpregs. *Mater. Manuf. Process.* **2021**, 1-10.
DOI:10.1080/10426914.2021.1885706.
- [8] Tucci, F.; Bezerra, R.; Rubino, F.; Carlone, P. Multiphase Flow Simulation in Injection Pultrusion with Variable Properties. *Mater. Manuf. Process.* **2020**, *35* (2), 152–162.
DOI:10.1080/10426914.2020.1711928.
- [9] Forcellese, A.; Simoncini, M.; Vita, A.; Di Pompeo, V. 3D Printing and Testing of Composite Isogrid Structures. *Int. J. Adv. Manuf. Technol.* **2020**, *109* (7–8), 1881–1893.
DOI:10.1007/s00170-020-05770-4.
- [10] Parmar, H.; Khan, T.; Tucci, F.; Umer, R.; Carlone, P. Advanced Robotics and Additive Manufacturing of Composites: Towards a New Era in Industry 4.0. *Mater. Manuf. Process.* **2021**, 1–35. DOI:10.1080/10426914.2020.1866195.
- [11] Vyavahare, S.; Kumar, S.; Panghal, D. Experimental Study of Surface Roughness,

- Dimensional Accuracy and Time of Fabrication of Parts Produced by Fused Deposition Modelling. *Rapid Prototyp. J.* **2020**, *26* (9), 1535–1554. DOI:10.1108/RPJ-12-2019-0315.
- [12] Ramezani Dana, H.; El Mansori, M.; Barrat, M.; Seck, C. A. Tensile behavior of additively manufactured carbon fiber reinforced polyamide-6 composites. *Polym.-Plast. Technol. Mater.* **2021**, DOI: 10.1080/25740881.2021.2005094
- [13] Verdejo de Toro, E.; Coello Sobrino, J.; Martínez Martínez, A.; Miguel Eguía, V.; Ayllón Pérez, J. Investigation of a Short Carbon Fibre-Reinforced Polyamide and Comparison of Two Manufacturing Processes: Fused Deposition Modelling (FDM) and Polymer Injection Moulding (PIM). *Mater.* **2020**, *13*, 672. <https://doi.org/10.3390/ma13030672>.
- [14] Dhakar, K.; Chaudhary, K.; Dvivedi, A.; Bembalge, O. An Environment-Friendly and Sustainable Machining Method: Near-Dry EDM. *Mater. Manuf. Process.* **2019**, *34* (12), 1307–1315. DOI:10.1080/10426914.2019.1643471.
- [15] Hart, N.; Brandon, N.; Shemilt, J. Environmental Evaluation of Thick Film Ceramic Fabrication Techniques for Solid Oxide Fuel Cells. *Mater. Manuf. Process.* **2007**, *15* (1), 47–64. DOI:10.1080/10426910008912972.
- [16] Bevilacqua, M.; Ciarapica, F. E.; Forcellese, A.; Simoncini, M. Comparison among the Environmental Impact of Solid State and Fusion Welding Processes in Joining an Aluminium Alloy. *Proc. Inst. Mech. Eng. Part B J. Eng. Manuf.* **2020**, *234* (1–2), 1–17. DOI:10.1177/0954405419845572.
- [17] Di Pompeo, V.; Forcellese, A.; Mancina, T.; Simoncini, M.; Vita, A. Effect of Geometric Parameters and Moisture Content on the Mechanical Performances of 3D-Printed Isogrid Structures in Short Carbon Fiber-Reinforced Polyamide. *J. Mater. Eng. Perform.* **2021**. DOI:10.1007/s11665-021-05659-7.
- [18] Kandananond, K. Optimization of Fused Filament Fabrication System by Response Surface Method. *Int. J. Metrol. Qual. Eng.* **2020**, *11* (4), 1-11. DOI:10.1051/ijmqe/2020002.

- [19] Chaidas, D.; Kechagias, J.D.; An investigation of PLA/W parts quality fabricated by FFF. *Mater. Manuf. Process.* **2021**, 1–9. DOI:10.1080/10426914.2021.1944193.
- [20] Jang, S.; Boddorff, A.; Jang, D.J.; Lloyd, J.; Wagner, K.; Thadhani, N. B.; Brettmann, B. Effect of material extrusion process parameters on filament geometry and inter-filament voids in as-fabricated high solids loaded polymer composites. *Addit. Manuf.* **2021**, *47*, 102313. DOI:10.1016/j.addma.2021.102313.
- [21] Zolfagharian, A.; Khosravani, M. R.; Kaynak, A. Fracture Resistance Analysis of 3D-Printed Polymers. *Polym.* **2020**, *12* (2), 302. DOI:10.3390/POLYM12020302.
- [22] Dul, S.; Fambri, L.; Pegoretti, A. High-Performance Polyamide/Carbon Fiber Composites for Fused Filament Fabrication: Mechanical and Functional Performances. *J. Mater. Eng. Perform.* **2021**, 1–20. DOI:10.1007/S11665-021-05635-1.
- [23] Haylock, R.; Rosentrater, K. A. Cradle-to-Grave Life Cycle Assessment and Techno-Economic Analysis of Polylactic Acid Composites with Traditional and Bio-Based Fillers. *J. Polym. Environ.* **2018**, *26* (4), 1484–1503. DOI:10.1007/s10924-017-1041-2.
- [24] Vita, A.; Castorani, V.; Germani, M.; Marconi, M. Comparative Life Cycle Assessment and Cost Analysis of Autoclave and Pressure Bag Molding for Producing CFRP Components. *Int. J. Adv. Manuf. Technol.* **2019**, *105* (5–6), 1967–1982. DOI:10.1007/s00170-019-04384-9.
- [25] Forcellese, A.; Marconi, M.; Simoncini, M.; Vita, A. Life Cycle Impact Assessment of Different Manufacturing Technologies for Automotive CFRP Components. *J. Clean. Prod.* **2020**, *271*, 122677. DOI:10.1016/j.jclepro.2020.122677.
- [26] Germani, M.; Mandolini, M.; Marconi, M.; Marilungo, E. A Method for the Estimation of the Economic and Ecological Sustainability of Production Lines. *Proc. CIRP* **2014**, *15*: 147–152. DOI:10.1016/j.procir.2014.06.072.
- [27] Halliwell, S. *End of Life Options for Composite Waste Recycle , Reuse or Dispose ?* National Composites Network: Chesterfield, Unite Kingdom. **2006**; 41.

- [28] Forcellese, A.; Marconi, M.; Simoncini, M.; Vita, A. Environmental and Buckling Performance Analysis of 3D Printed Composite Isogrid Structures. *Proc. CIRP*. **2021**, *98*, 458–463. DOI:10.1016/j.procir.2021.01.13.
- [29] Gervásio, H.; Silva, L. S. da. Comparative Life-Cycle Analysis of Steel-Concrete Composite Bridges. *Struct. Infrastruct. Eng.* **2008**, *4* (4), 251–269. DOI:10.1080/15732470600627325.
- [30] Bianchi, I.; Forcellese, A.; Marconi, M.; Simoncini, M.; Vita, A.; Castorani, V. Environmental Impact Assessment of Zero Waste Approach for Carbon Fiber Prepreg Scraps. *Sustain. Mater. Technol.* **2021**, *29*, e00308. DOI:10.1016/j.susmat.2021.e00308.
- [31] Garcia, F. L.; Nunes, A. O.; Martins, M. G.; Belli, M. C.; Saavedra, Y. M. B.; Silva, D. A. L.; Moris, V. A. da S. Comparative LCA of Conventional Manufacturing vs. Additive Manufacturing: The Case of Injection Moulding for Recycled Polymers. *Int. J. Sustain. Dev. Plan.* **2021**, 1–9. DOI:10.1080/19397038.2021.1990435.
- [32] Yosofi, M.; Kerbrat, O.; Mognol, P. Energy and Material Flow Modelling of Additive Manufacturing Processes. *Virtual. Phys. Prototyp.* **2018**, *13* (2), 83–96. DOI:10.1080/17452759.2017.1418900.
- [33] Ribeiro, I.; Kaufmann, J.; Götze, U.; Peças, P.; Henriques, E. Fibre Reinforced Polymers in the Sports Industry – Life Cycle Engineering Methodology Applied to a Snowboard Using Anisotropic Layer Design. *Int. J. Sustain. Eng.* **2018**, *12* (3), 201–211. DOI:10.1080/19397038.2018.1508318.