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Development of an Additive Manufacturing System for the Deposition of Thermoplastics Impregnated with Carbon Fibers [†]

Miguel Reis Silva [1,2,*], **António M. Pereira** [1], **Nuno Alves** [1], **Gonçalo Mateus** [3], **Artur Mateus** [1] **and Cândida Malça** [1,4]

[1] Centre for Rapid and Sustainable Product Development, Polytechnic Institute of Leiria, Rua de Portugal, 2430-028 Marinha Grande, Portugal; mario.pereira@ipleiria.pt (A.M.P.); nuno.alves@ipleiria.pt (N.A.); artur.mateus@ipleiria.pt (A.M.); candida@isec.pt (C.M.)

[2] Institute for Polymers and Composites/I3N, University of Minho, Campus of Azurém, 4800-058 Guimarães, Portugal

[3] Plastimago—Transformadora de Plásticos, Lda., Estrada de Leiria, 2430-091 Marinha Grande, Portugal; goncalo@plastimago.pt

[4] Department of Mechanical Engineering, Polytechnic Institute of Coimbra, Rua Pedro Nunes, 3030-199 Coimbra, Portugal

[*] Correspondence: miguel.r.silva@ipleiria.pt; Tel.: +351-244-569-441

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Abstract: This work presents an innovative system that allows the oriented deposition of continuous fibers or long fibers, pre-impregnated or not, in a thermoplastic matrix. This system is used in an integrated way with the filamentary fusion additive manufacturing technology and allows a localized and oriented reinforcement of polymer components for advanced engineering applications at a low cost. To demonstrate the capabilities of the developed system, composite components of thermoplastic matrix (polyamide) reinforced with pre-impregnated long carbon fiber (carbon + polyamide), 1 K and 3 K, were processed and their tensile and flexural strength evaluated. It was demonstrated that the tensile strength value depends on the density of carbon fibers present in the composite, and that with the passage of 2 to 4 layers of fibers, an increase in breaking strength was obtained of about 366% and 325% for the 3 K and 1 K yarns, respectively. The increase of the fiber yarn diameter leads to higher values of tensile strength of the composite. The obtained standard deviation reveals that the deposition process gives rise to components with anisotropic mechanical properties and the need to optimize the processing parameters, especially those that lead to an increase in adhesion between deposited layers.

Keywords: additive manufacturing; fused filament fabrication; oriented extrusion; reinforced composites; carbon fibers; prepregs yarns; thermoplastic composites; polyamide

1. Introduction

Composite materials, which are obtained by the combination of different materials, exhibit distinct properties, usually superior to those of the constituents they gave rise to. Currently, composite materials are produced for structural purposes due to their high elastic modulus and mechanical strength in the most diverse industries including aeronautics, aerospace, nautical, canoeing, sporting goods, consumables, infrastructure, among others [1–3]. Gaps in production methods have limited

the widespread use of composite materials for other purposes. Recently, processing methods such as pultrusion, resin transfer molding (RTM), structural reaction injection molding (SRIM), compression molding compounding (SMC) and filament winding have been automated to achieve high production rates [4–8]. Some of the techniques are restricted to the use of thermosetting resins while others may use both thermoplastic and thermosetting resins. Some of these techniques may use the continuous fibers as reinforcement while others are restricted to the use of short fiber reinforcements [4–10]. The processing of the thermoplastic matrix composites reinforced with long or continuous fibers causes constraints on the impregnation of the fibers due to the high viscosity that the thermoplastic matrices present [10]. However, the interest in composites reinforced with continuous thermoplastic matrix fibers has been increasing significantly due to advances in fiber impregnation. Thus, in recent years the development of impregnation technologies has made pre-impregnated systems with adequate quality available, in order to make composites with more interesting and reproducible mechanical properties with the different thermoplastic matrix/fiber combinations [10].

From manual deposition to actual fabrication through automatic deposition and consolidation, composite manufacturing technology has progressed rapidly thanks to the additive manufacturing processes [11–13]. Additive manufacturing (AM) technology can be used in composite processing to produce complex geometric structures and components from 3D models with almost no material waste [11,12]. In addition, AM technology allows one to take advantage of the directional stiffness properties of the composites, i.e., the fibers must be deposited and oriented layer by layer according to the orientation and trajectory that optimizes their performance, namely in terms of stiffness and resistance according to the desired application [14–16]. The automatic deposition of continuous fibers in thermoplastic matrix has existed for about 25 years, and several companies have developed devices for the deposition and consolidation of continuous fibers in thermoplastic matrix, mainly through robotic arms. However, because of their complexity, these devices have a very high acquisition cost and are prohibitive for small and medium sized industries. Thus, the study and development of new systems for the deposition and consolidation of continuous fibers in a thermoplastic matrix is assumed to be a pertinent and current challenge. In this work, a system of deposition and consolidation of a filament of continuous fibers or long fibers in a thermoplastic matrix, according to the predefined orientations that comply with the service requirements of the component, is shown as an extension of a previous work presented by the authors [17]. This innovative system allows the usage of low-cost 3D printers (i.e., with slight modifications) for the production, with hybrid filament of fiber blend, of components in composite material of polymer matrix, contributing to the increase of the competitiveness of the products aimed at engineering applications.

1.1. Processing of Composite Reinforced with Fibers

Thermoplastic matrix composites provide many advantages when compared with thermosetting matrix composites. Some examples of these advantages are: higher fracture and deformation resistance, shorter processing cycle time, no extra time required for the polymerization reaction to occur, long service life without the need for refrigeration at negative temperatures, good chemical resistance (solvents), high potential for the application in the manufacturing process, as well as the fact that they can be recycled. However, molten thermoplastics have a high viscosity (500–5000 Pa·s) compared to thermosetting (typically 100 Pa·s), which makes their processing more problematic [18]. This drawback results in the difficulty of impregnation of the reinforcing fibers, in porosity of the products and, possibly, in low and unpredictable mechanical performance. It is precisely at this point that processing-related aspects take on crucial importance. The development of new deposition and consolidation strategies is the key to overcoming these constraints. One of the important challenges lies in the processing knowledge, which in recent years has gone from practices based on empirical knowledge to procedures based on scientific and engineering principles. These processing difficulties require novel processing techniques involving pre-impregnated tapes and hybrid filaments of reinforcing fibers/matrices. The knowledge related to the processing of these types of materials assumes an increasing importance

in the current context of globalization and high competitiveness between companies, where efficiency and speed of production are determinant factors for success. As previously mentioned, thermoplastic matrix composites present promising characteristics for the composite industry. Its post-processing and the possibility of reprocessing are, in addition, characteristics that demonstrate the potential cost reduction that these materials can represent [19,20].

The mechanical properties of the composites are strongly dependent on the type of reinforcement fibers and, of course, their properties. The matrix/fiber interface is also determinant in the final properties of the composite because only a proper adhesion will allow for a suitable response to the requests to which the component being used is subject. The most common reinforcing fibers are glass, Kevlar and carbon fibers (CF) [21]. Currently, carbon fibers are the most used for high performance applications, namely in the industries of aeronautics, aerospace, automobile, among others [2,13,22–25]. This is the reason why they were selected to carry out this study. The relationship between the length and the mean diameter of the short fibers is called the size factor. A composite of short fibers is strongly dependent on this size factor, i.e., the larger the size factor, the greater the composite strength. The carbon fibers can be classified according to their mechanical properties, as shown in Table 1, or according to the precursor used in their manufacturing.

Table 1. Carbon fibers properties [25].

Type of Fiber	Modulus of Elasticity [MPa]	Tensile Strength [MPa]	Deformation at Break [%]	Thermal Expansion Coefficient [10^{-6}/°C]	Density [g/cm^3]
PAN	200–400	2480–5600	0.6–1.2	−0.7 to −0.5	1.8

The combination of the matrix and the reinforcement prior to the production of the component is called prepreg. In the case of thermoplastic matrices, this operation produces a thin tape of unidirectional continuous fibers, impregnated with polymer with the volume fraction of the desired fibers. The result of this operation is a rigid and a non-sticky material. The obtained tape may be employed in the manufacture of the components having the desired geometries by means of compression molding, autoclaving or application of tape deposition techniques [26]. This combined method allows for good control of the orientation of the fibers, as well as a precise fiber content, in addition to guaranteeing a uniform distribution of the fibers. The resulting products are completely consolidated, i.e., without macroscopic voids. All these characteristics represent high-quality composites with consistent mechanical properties. However, the appearance of this rigid and non-sticky composite thermoplastic tape implies difficulties and increased processing costs, limiting its application to high performance products.

The pre-impregnated hybrid filaments consist of the intimate blend of reinforcing filaments with filaments of the thermoplastic matrix, at the volume fraction of desired fibers [27]. This form of prepreg has a high fiber blend (matrix and reinforcement), which results in a reduction, in the order of micrometers, of the impregnation distance—the length that the molten matrix must travel to envelop reinforcement fibers. This form of prepreg also makes it possible to take full advantage of cost reduction in the production of prepregs and allows for a wide choice of different matrices and reinforcement combinations. Various techniques are used in the production of hybrid filaments by reinforcing/matrix fiber blending [28]. One technique is to use discontinuous filaments (stretch-broken) with a predetermined length, which are twisted into a yarn. The average fiber length, around 100 mm, is considerably longer than the critical fiber length, i.e., the use of long reinforcing fibers with a length of 100 mm does not influence the longitudinal modulus of the composite as compared to the use of continuous fibers [29]. However, due to the imperfection of fiber alignment resulting from the use of twisted filaments, the longitudinal properties of the composites may be affected. Even so, the interlayer properties may be reinforced because of the misalignment of the fibers according to the interlaminar directions, thus increasing mechanical properties [30].

In this work a commercially available commingled yarn, made of mixed CF and polyamide 12 fibers (PA12 fibers) under trade name TPFL® produced by Schappe Techniques (France) and 1.75 mm

polyamide 12 (PA12) filament supplied by eSUN (China) was selected. The selection of this material refers to the advantages listed above.

1.2. Additive Manufacturing by Filamentary Fusion

Fused filament fabrication (FFF) was invented by Stratasys Inc. founder Scott Crump, and it emerged in the 1980s [31]. The FFF technology-built components are produced layer by layer, from bottom up, by heating and extruding thermoplastic filaments. The automatic deposition systems of thermoplastic matrix pre-impregnated fibers are production processes that do not require an autoclave, which represents, especially in the aeronautics sector, a competitive advantage in the cost of high-performance composites [31–33]. The deposition of pre-impregnated fibers into thermoplastic matrices removes the costs of the pre-impregnation, the cold storage and autoclave procedures from the process. It also facilitates the production of high-dimension structures with complex layups, as it does not require a maximum processing time by impregnation [34]. The existing equipment for the additive manufacture of components in thermoplastic matrix composites can be divided into two groups:

i. the first group is comprised of customized, technologically complex equipment, and developed by a small group of companies with capacity and technological knowledge of the process. This kind of equipment allows for the 3D deposition of fibers and is used in the production of medium/large-sized components with a high cost, for applications of high performance, such as aeronautics and aerospace areas [35,36];

ii. the second group consists of small filamentary fused additive manufacturing equipment with the capability to process 2D fibers in a plane; the number of manufacturers of this class of equipment is even smaller, with the current commercial offer being limited to a single manufacturer. This kind of equipment has the capacity to produce small components at a reduced cost for medium performance applications [37].

It is in the second group of equipment that the system presented in this paper fits. Currently, the commercial offer of equipment which is less complex and with reduced dimensions is limited to a single company: Markforged. The most recently developed equipment, the Mark Two printer, presents the printing of carbon, aramid and glass fibers in a polyamide matrix as its main characteristic. This equipment uses a filament composed of continuous fibers inside, encased in polyamide. The printer uses two direct extrusion heads: one for the filament with fibers and another for the polymer filament. The Markforged technology uses proprietary filaments of pre-consolidated continuous fibers in a thermoplastic matrix, which allows the filament to be drawn and compressed normally by the extrusion rolls. The approach proposed here by the authors uses a commercially available commingled yarn, made of mixed CF and polyamide fibers (PA12 fibers), which is not rigid enough to be compressed by the extrusion rollers and allows their passage through the extrusion nozzle. This approach uses a flow of molten polymer capable of generating shear stresses on the surface of the fibers, thus promoting their displacement to the outlet of the heated nozzle [37].

Recently, the Anisoprint announced the development of an FFF printer, capable of printing traditional thermoplastics with carbon fiber reinforcement. It is indisputable that high performance components manufacturing intended for engineering applications is strongly valued by the incorporation of fiber reinforcements. The advantages that would result from the combination of the current FFF additive manufacturing system with a continuous and non-continuous fiber deposition and consolidation system, which would make the additive manufacture of thermoplastic matrix composites an economically feasible manufacturing process would also be undeniable [38]. This enhanced manufacturing technology would make it possible to reinforce the components only in the regions and in relation to the most requested directions according to their application in use. This feature can be achieved using reduced diameter yarn filaments, which will be deposited solely according to the most requested orientations, allowing it to produce micro reinforcements, as well

as larger areas of reinforcement [39]. With this purpose, a prototype system was designed, built and tested and it is presented in the next section.

2. Materials and Methods

2.1. Fiber Deposition System

In this work, a long fiber deposition system for integration in 3D printers recently developed by the authors [17] is presented. The development of this system was based on the concepts and technology developed by the RepRap movement, founded in 2005 by Dr. Adrian Bowyer. The FFF prototype equipment was constructed from extruded aluminum profiles and an ISEL brand XYZ axis system with trapezoidal spindle movement, as shown in Figure 1.
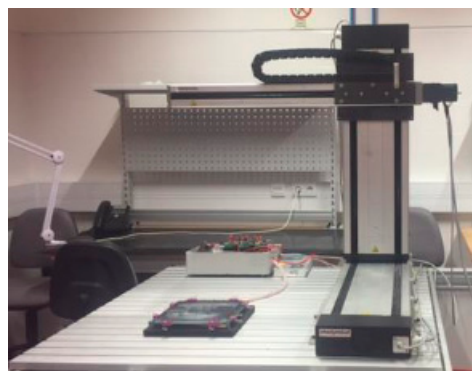
Figure 1. Fused filament fabrication (FFF) prototype equipment.

The equipment systems of control and monitoring were developed using an Arduino Mega 2560 controller, 3 external drives MSD542-V2.0 and in NEMA 23 step motors, as shown in Figure 2. As both the hardware and the adopted software are open source, the FFF prototype developed can be considered a low-cost system. The development of this coextrusion system is intended to promote the melting of the polymer fibers present in the TPFL® filament and their controlled deposition.
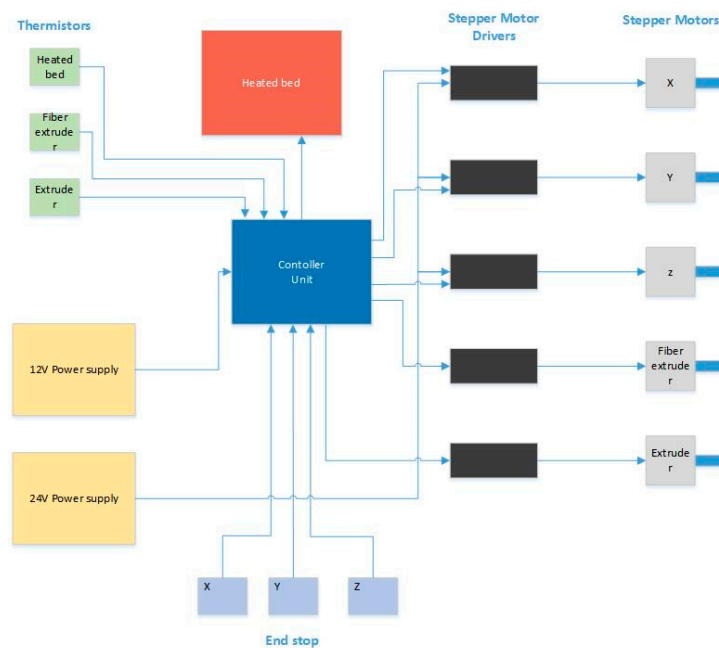

Figure 2. FFF prototype equipment electronic diagram.

Since the carbon fiber filaments mixed with PA12 do not have enough stiffness to be compressed by the extrusion rollers, enabling their passage through the heated nozzle, it is necessary to use a flow of molten polymer capable of generating shear stresses on the surface of the fibers, thus promoting its displacement/drag towards the heated nozzle outlet. The strategy selected to join the fiber filament with the thermoplastic filament was to feed the fiber filament before thermal cutting in a cold zone, where the polymer is still in a solid state, as shown in Figure 3, detail b).
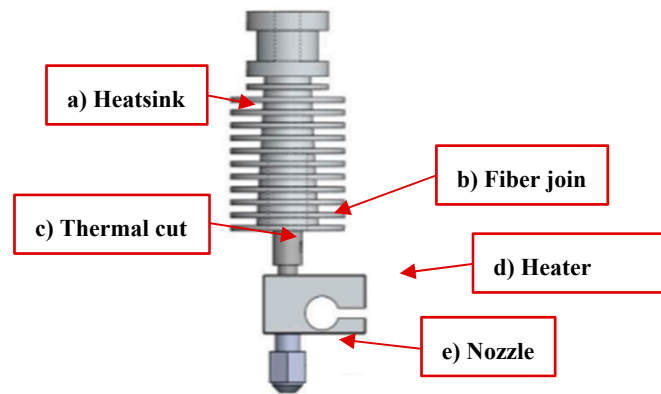

Figure 3. Deposition nozzle system; (**a**) heatsink zone; (**b**) fiber join hole; (**c**) heat break (thermal cut); (**d**); heater block; (**e**) extrusion nozzle.

The developed heat break component has as its function to: (i) feed the fibers to the thermoplastic filament and (ii) allow the thermal cut between the hot zone and the heatsink to avoid the melting of the polymer outside the heated zone which would cause clogging in the system. This component must, additionally, guarantee the maintenance of the hot zone temperature, by minimizing heat transfer by conduction, and finally allow for the protection of the polymeric components and the adjacent electronic systems. The material selected was stainless steel AISI 316L, which is a refractory steel, and has a low coefficient of thermal conductivity of 16.3 W/m·K at 100 °C.

The developed heat break described above was produced using a SLM Selective Laser Melting (SLM Solutions SLM 125hl model) metal additive manufacturing equipment. The metallic powder used was 316L stainless steel supplied by SLM Solutions. The selection of this AM technique allowed for manufacturing reduction time of the different prototype parts by the simultaneous production of the same component with slight variations of geometry. However, the SLM requires pre- and post-processing operations and holes rectification. Figure 4 shows the components produced, still fixed to the construction platform by the supports, prior to carrying out the post-processing operations.

Figure 4. Components produced by SLM prior to performing the post-processing.

The solution adopted allows the introduction of the fiber filament into the extrusion channel before the thermal cut of the extrusion system, as shown in Figure 5.
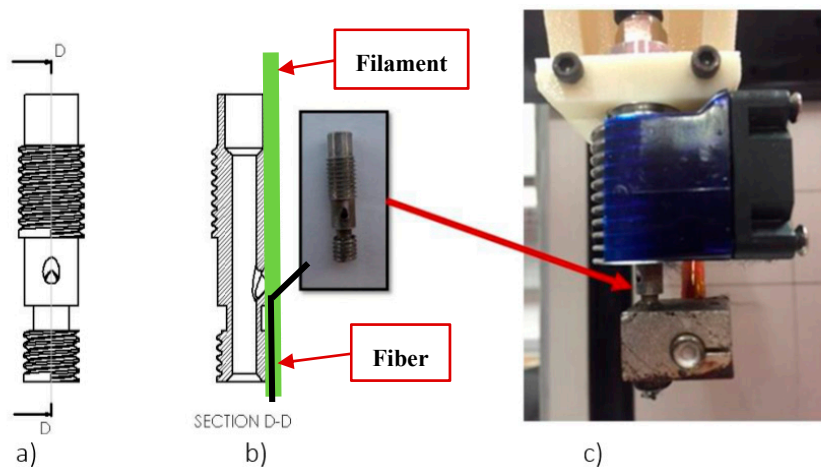
Figure 5. Fiber deposition nozzle with thermal cut detail: (**a**) main view; (**b**) cut view showing filament and fiber yarn; (**c**) real deposition head.

In addition, a fiber-cutting system was developed and applied at the outlet of the heated nozzle. This system allows for the automatization of the fiber-cutting operation. The cutting of the fibers is performed by a diamond disc coupled to a direct current (DC) motor. The motor backing is coupled to a hinged mechanism that allows for the reciprocating movement of the disc between the fiber cutting position and the standby position. The drive mechanism of the disc is composed of a NEMA 8 stepper motor, a V-spindle, a connecting rod and a guiding assembly consisting of a chute and bushings, as shown in Figure 6. Since it is a prototype of a device, the polymeric materials selected for the construction of the chassis, connecting rod and bushes were ABS and PA6, respectively. In the manufacture of these components, a FFF polymer additive manufacturing equipment was used to model uPrintSE from the Stratasys. The cutting system is triggered at the end of the deposition of each layer of fiber. This action is controlled by the inclusion of a routine in the Gcode file of the component manufacture. This routine allows for the deposition pause by moving the printhead vertically, thus activating the cutting mechanism. After cutting, the disc collects and the deposition of the fibers starts in the next layer, as exemplified by the sequence of images in Figure 6. During this routine the temperature control of both the extrusion and the deposition layer is maintained.
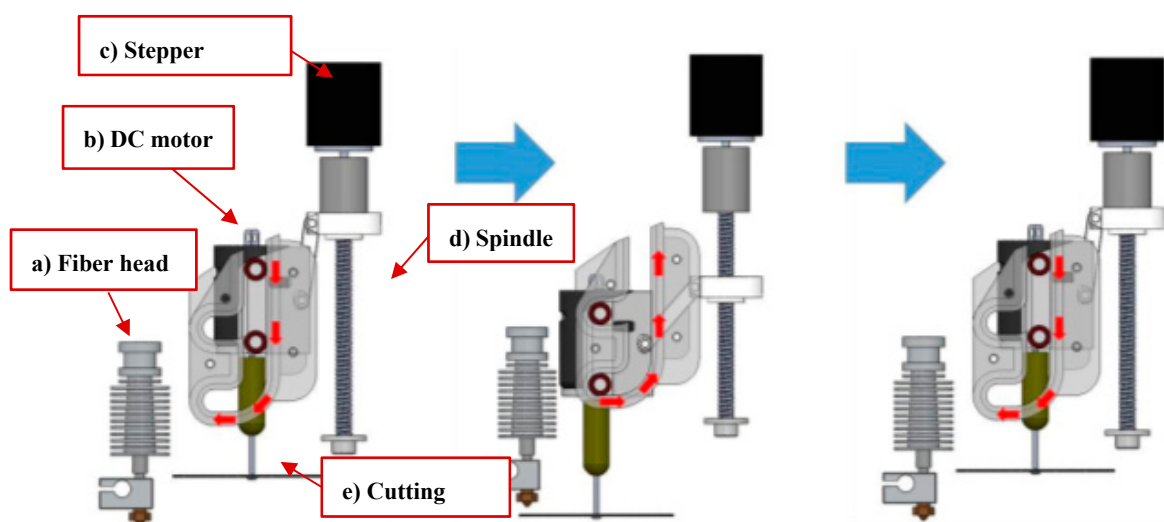

Figure 6. Operating scheme of fiber cutting system; (**a**) fiber deposition head; (**b**) direct current (DC) motor; (**c**) stepper motor; (**d**) spindle; (**e**) fiber cutting disc.

In order to integrate the two extrusions and cutting systems, a specific support was developed and applied to the X-axis carriage of the printing equipment. The geometry of this support and its dimensions have been designed, considering the specific system of the axes used. Figure 7 shows the assembly of the system. The arrangement of the axle system and the high dimensions caused constraints, preventing a greater compactness of the support. This support should therefore be reengineered with a purpose of its integration into small FFF systems. For the extrusion of PA12, a commercially available 1.75 mm E3D-v6 extrusion system was used. This extrusion system fulfills the exact specifications required using PA12, namely the high melting temperatures of the polymer (~300 °C) and its geometry to avoid clogging [40].
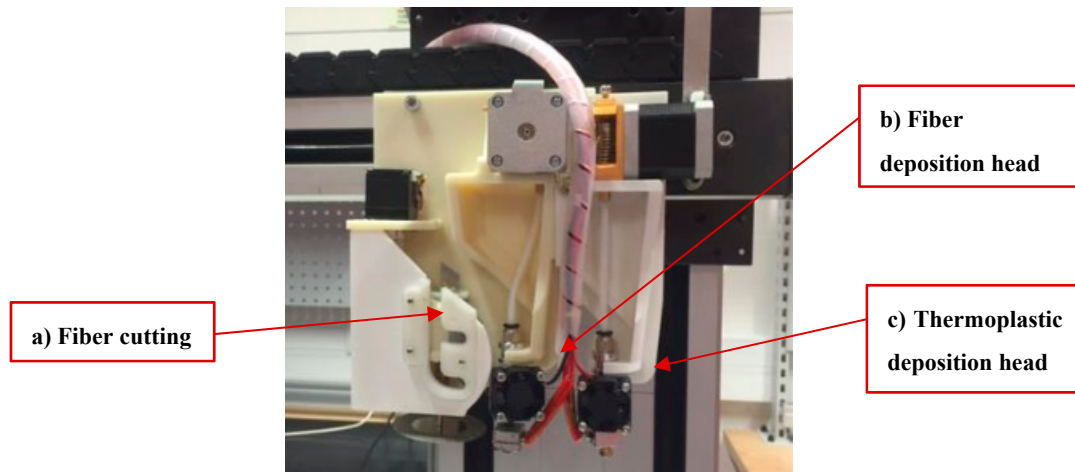

Figure 7. Integrated double extrusion and fiber cutting systems; (**a**) fiber cutting system; (**b**) fiber deposition head; (**c**) thermoplastic filament deposition head.

Adjustments and calibrations were made to the system prior to the functioning of the developed printing system. The first procedure, after loading the firmware in the controller of the equipment, was the verification of the axes' direction of movement and the logic of the signal of the limit switches according to the firmware. Then, the maximum displacement length of the three axles was calculated and the construction platform leveled. Finally, printing tests were performed to adjust deposition parameters such as velocity, acceleration and its derivative (jerk). It was also necessary to adjust the percentage of polymer added to the fiber strand in the coextrusion.

The composite additive manufacturing, through the FFF technology needs to respect and adapt the procedures to overcome the limitations of the materials and the process, aiming to optimize the mechanical properties of the components produced. To pursue this objective, some rules were followed from the modeling phase of the components to the preprocessing of the file generation of the trajectories, as well as the processing parameters, such as the processing temperature, the deposition velocity, and the height and width of the layers. Table 2 summarizes the values of the processing parameters assumed after an optimization process. Samples with dimensions and geometries, according to the standards required, were printed for to carry out tensile and three-point bending tests. In the following section, these and other tests are carried out with the aim of characterizing printed composites.

Table 2. Processing parameters.

Processing Parameters	Units	Value
Construction platform temperature	°C	110
Deposition temperature	°C	270
Deposition speed	mm/s	5
Nozzle inner diameter	mm	1.5
Layer height	mm	1
Layer width	mm	1.5

2.2. Materials

In this work the additive manufacturing of reinforced thermoplastic composites was carried out using a 1.75 mm polyamide 12 (PA12) filament supplied by eSUN and a hybrid filament of carbon fibers (CF) and polyamide 12 (PA12), TPFL® by Schappe Techniques (France). The TPFLs are dry prepregs that homogeneously combine reinforcement and matrix filaments. This filament combines long CF and PA12. The matrix is composed of PA12 produced by the company EMS CHEMIE AG (Switzerland) and it is marketed under the name Grilamid 12. The selection of this matrix took into consideration criteria such as adhesion, processing temperatures, performance, durability and cost. The properties of the matrix are shown in Table 3. The PA12 is a semi-crystalline thermoplastic polymer with a crystallinity of about 41% for solidification rates between 1 and 100 °C/min [27].

Table 3. Properties of Grilamid 12 [27].

Density [g/cm^3]	Melting Viscosity [Pa·s]	Glass Transition Temperature [°C]	Melting Temperature [°C]	Elasticity Modulus [MPa]
1.01	250	42	178	1100

For the hybrid filament manufacturing, discontinuous PA12 fibers which have a mean diameter of 20 μm are used. The reinforcement of the composite material used in this work is composed of long CF supplied by Toho-TenaxFibers GmbH (Germany). Carbon fibers are produced from continuous fiber filaments which are drawn by means of rollers to promote their breakage in distinct sections. This technique allows for the elimination of the weakest points of the fibers and for the improvement of their characteristics, increasing their tenacity and spinning capacity. Thereafter, the long fiber filaments are blended with the PA12 fibers, and may contain 1000 or 3000 carbon filaments, which are defined as 1 K and 3 K, respectively. This mix is carried out to keep the carbon and PA12 fibers parallel to each other and oriented towards the filament. According to Schappe Techniques the fiber volume fraction including the outer winding is 53% with the metric number of 0.84 Mc (km/kg). Some properties of the TPFL® filament are shown in Table 4.

Table 4. Properties of TPFL® hybrid filament [27].

Properties	Units	1 K Yarn	3 K Yarn
Total volume fraction of carbon fibers, Vfc	%	55	55
Length of carbon fibers, Lfc	mm	50	200
Length of PA12 fibers, Lfm	mm	75	85
Density of carbon fibers, ρc	g/cm^3	1.77	1.77
Density of matrix, ρm	g/cm^3	1.01	1.01
Diameter of carbon fibers, Dfc	μm	5	7
Diameter fibers of PA12, Dfm	μm	20	20

The methods used to obtain the composite mechanical characterization and the adhesion between yarns—in the same layer and between yarn layers—were both uniaxial tensile tests, and the three-point bending/flexural tests were performed. The fiber deposition path was made in longitudinal from

the middle of the sample to the outer region. After the production, the test samples were manually polished with 220 grit sandpaper to eliminate burrs and edges and to standardize their dimensions. The specimens were printed at an ambient temperature of 23 °C. Polyamide filament coils and carbon fibers/PA12 yarn, prior to use, were kept in an oven for 5 h at a temperature of 70 °C. This procedure aimed to eliminate the moisture present in the polyamide [41]. The uniaxial and three-point bending flexural tensile tests were performed on a Zwick electrochemical test equipment, model Z100. The specimens were taken to the breaking point and the data obtained was analyzed and processed.

The calculation of longitudinal modulus (E_1) of composites reinforced with fibers were theoretically determined and the values obtained are presented in Table 5. For the reinforcement with unidirectional fibers, Equation (1) can be applied to calculate E_1 [29]:

$$E_1 = E_F V_{fc} + E_M \left(1 - V_{fc}\right) \tag{1}$$

where, E_F and E_M are the modules of carbon fibers and PA12 matrix, respectively, and V_{fc} is the carbon fiber volume fraction. For the reinforcement with long fibers, the longitudinal modulus (E_{1disc}) can be estimated using Equation (2) [29]:

$$E_{1disc} = \frac{E_M \left(1 + \xi \eta V_{fc}\right)}{1 - \eta V_{fc}} \tag{2}$$

where, ξ and η are given by Equation (3):

$$\xi = 2 \left(\frac{L}{d_F}\right); \qquad \eta = \frac{\left(\frac{E_F}{E_M}\right) - 1}{\left(\frac{E_F}{E_M}\right) + \xi} \tag{3}$$

Table 5. Young's modulus calculation results (c) the authors, CC BY-NC-ND 4.0 [17].

CF/PA12 Yarn	Units	1 K Yarn	3 K Yarn
Young modulus of the matrix, E_M	GPa	1.1	1.1
Young modulus of the carbon fiber, E_F	GPa	240	240
Carbon fiber volume fraction, V_{fc}	1	0.0345	0.1173
Carbon fiber average length, L_F	mm	100	100
Carbon fiber diameter, D_F	μm	7	7
Young modulus of composites (Continuous CF), E_1	GPa	9.34	29.12
Young modulus of composites (Long CF), E_{1disc}	GPa	9.28	28.93
ξ	1	28,571.4	28,571.4
η	1	0.007544	0.007544

From the results of Table 5 and for the 3 K yarn, the value of volume fraction (V_{fc}) and the diameter of carbon fibers (D_F) are 0.1173 and 7 μm respectively, whereas the longitudinal elasticity modules of carbon fibers and PA12 (E_F) = 240 GPa and (E_M)= 1.1 GPa with an average length of carbon fibers being 100 mm. The theoretical calculation of the longitudinal module, through the Equations (1) and (2), is 29 GPa in both cases. Regarding the 1K yarn, the value of volume fraction (V_{fc}) and the diameter of carbon fibers (D_F) is 0.0345 and 7 μm respectively, whereas the longitudinal elasticity modules of carbon fibers and PA12 (E_{fc}) = 240 GPa and (E_M) = 1.1 GPa with an average length of carbon fibers being 100 mm. The theoretical calculation, through the Equations (1) and (2), is around 9 GPa in both cases. These results mean that, theoretically, although the carbon fibers are discontinuous, the mechanical properties are like those of composites reinforced with continuous fibers. This is due to the high aspect ratio—the ratio between length and diameter—exhibited by the carbon fibers.

2.2.1. Uniaxial Tensile Tests

The tensile tests were performed only for a four-layer sample with 3 K yarn, the dimensions of the test samples were selected according to the maximum build dimensions of the prototype equipment. Figure 8 illustrates the geometry and dimensions of the uniaxial test sample used called the "dog bone". The average thickness obtained in the test pieces was 4 mm. The uniaxial tensile tests were performed in position control until the specimen rupture, with a constant displacement velocity of 1 mm/min and a preload of 2 N, at room temperature. Three tests were carried out to minimize possible interferences resulting from the processing and testing of test pieces.
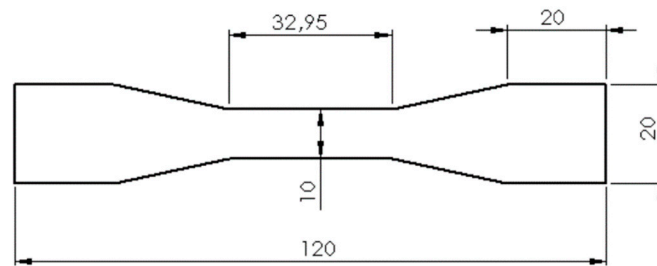
Figure 8. Dog bone configuration sample, dimensions in mm (c) the authors, CC BY-NC-ND 4.0 [17].

2.2.2. Three-Point Bending Tests

The three-point bending test was performed according to ASTM D790 [42]. The specimens were cut according to a rectangular geometry with the dimensions of 60×15 mm, as shown in Figure 9a and later sanded with 220 grain sandpaper to eliminate burr and surface softening, as shown in Figure 9b. The thickness and average width of each specimen were then checked. Three samples each of thickness 2 and 4 mm were tested and, for the two types of yarn 1 K and 3 K, the average was then calculated to minimize possible human or material faults.
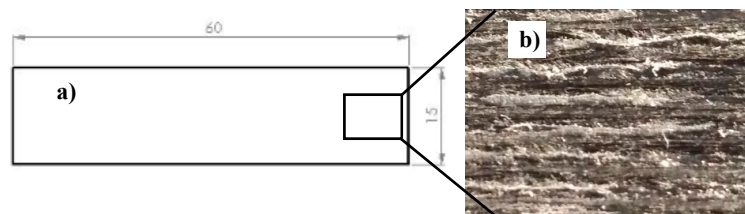

Figure 9. Three-point bending sample; (**a**) sample drawing, dimensions in mm; (**b**) fiber orientation detail (c) the authors, CC BY-NC-ND 4.0 [17].

2.2.3. Morphological and Microstructural Analyses

Scanning electron microscopy (SEM) (Merlin-61-50 FE-SEM, from Carl Zeiss) was used to characterize the microstructure. The cross-sectional images were used to demonstrate the distribution of the long fibers, any possible void content, and the bonding details. The surface of the SEM samples was first sputter coated with gold with a thickness of 20 nm. Then, SEM micrographs were observed at an acceleration voltage of 5 kV and an emission current of 20 pA.

3. Results and Discussion

This section provides a concise and precise description of the experimental mechanical and SEM test results, and their interpretation as well as the experimental conclusions that can be drawn.

3.1. Uniaxial Tensile Tests

From the uniaxial tensile tests results, it is possible to conclude that a rupture stress of 153.62 MPa with a standard deviation of 5.16% was obtained for 3 K four-layer specimens. Figure 10 shows the

curve stress-strain obtained for the 3 K yarn. The mean displacement at break was 2.12 mm with a standard deviation of 0.61%. The tensile strength is found to have a lower value according to the low density of carbon fibers present in the composite. Regarding its standard deviation, it is within the expected, due to the deposition process that originates in components with high anisotropic mechanical properties, according to the manufacturing directions. Another factor responsible for this standard deviation can be related to processing parameters, which need optimization.
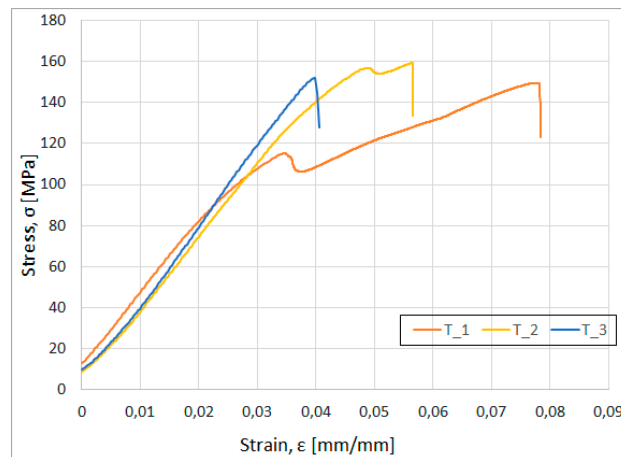
Figure 10. Results of uniaxial tensile test for the 3K yarn with four layers (c) the authors, CC BY-NC-ND 4.0 [17].

3.2. Three-Point Bending Tests

Figure 11 depicted the results of the three-point bending test for the 3 K yarn and Table 6 summarizes the results of the three-point bending tests performed. It is possible to conclude from Table 6 that regarding the three-point flexural results with the increase of yarn layers, from 2 to 4, the break strength increased by about 366% and 325% for 3 K and 1 K yarn, respectively. This increase was essentially due to the greater number of fibers, which is around 100%. For the 3 K yarn, there was an increase of approximately twice the tensile break value than those obtained with 1 K yarn. This is essentially due to the increase of the fiber yarn diameter in the 3 K specimens. With the decrease of the amount of polymer added, the percentage of fibers is higher, which leads to a high tensile strength value of the composite.
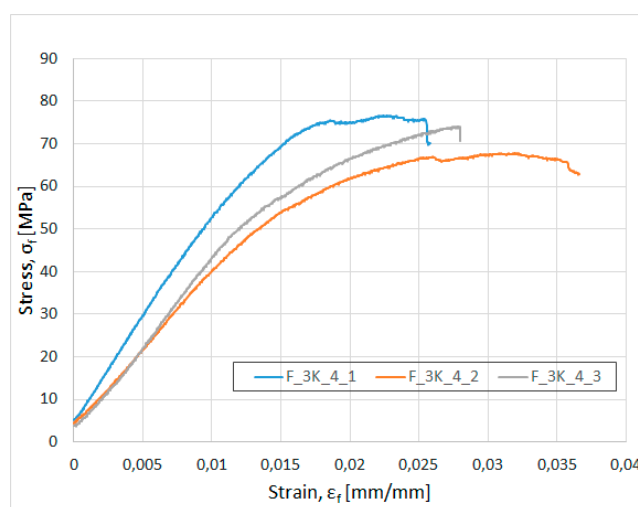

Figure 11. Results of three-point bending test for 3K yarn with four layers (c) the authors, CC BY-NC-ND 4.0 [17].

Table 6. Three-point bending test data (c) the authors, CC BY-NC-ND 4.0 [17].

Sample Reference	B [mm]	H [mm]	Maximum Force [N]	Ultimate Strength [MPa]	Mean Ultimate Strength [MPa]	Standard Deviation [MPa]
F_3K_2_1	13.6	2	84.6	74.6		
F_3K_2_2	14.3	2	93.0	78.0	81.3	8.7
F_3K_2_3	14.3	1.6	69.6	91.2		
F_3K_4_1	14.3	3.7	312.4	76.6		
F_3K_4_2	13.9	4	314.3	67.8	71.2	4.9
F_3K_4_3	14.1	3.5	266.6	74.1		
F_3K_4_4	14.2	4	313.7	66.3		
F_1K_2_1	14.5	2.4	77.4	44.5		
F_1K_2_2	14.4	2.3	62.0	39.1	42.8	3.2
F_1K_2_3	14.5	2.3	71.8	44.9		
F_1K_4_1	14.2	3.8	217.5	50.9		
F_1K_4_2	14.2	4	224.0	47.3	48.5	2.6
F_1K_4_3	14.2	4.2	246.2	47.2		
F_PA_4_1	15	4.4	582.2	96.2		

The standard deviation obtained reveals a need to optimize the adhesion between layers, which means that the processing parameters (i.e., temperature and layer thickness) must be optimized. Figure 11 shows that, after the rupture tension is reached, there are rebounds of resistance decrease, i.e., a break by levels is verified. This behavior may be due to the occurrence of a progressive inter-filament rupture, i.e., to the rupture of adhesion between filaments. To explore this issue, scanning electron microscopy (SEM) observations were made and they are presented in the next section.

3.3. Scanning Electron Microscopy (SEM) Results

To perform morphological and microstructural analyses of processed composites and to analyze the modes of failure of the tested samples in detail, before and after mechanical tests, tensile and bending tests, SEM observations were made. Figures 12–14 show SEM images of the transversal section of the sample printed using the 1 K and 3 K yarn before and after mechanical tests, respectively. For the tensile test, the failure mode developed was the interfilamentar long splitting while for the bending case only the interfilamentar failure is observed. Both Figures 12b and 13b show that the composite is not completely consolidated, due to the voids that can be observed in Figures 13a and 14a,b. In addition, the impregnation distance is high. Figure 12b presents a planar surface of PA12 matrix and clear interface with the fiber bundle.
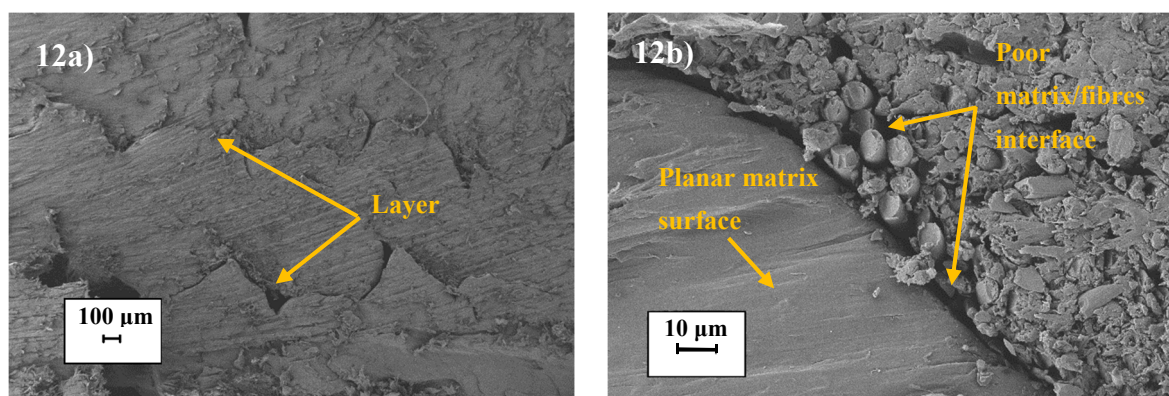

Figure 12. 1 K fibers sample before mechanical tests; (**a**) layer structure; (**b**) matrix/fibre interface.
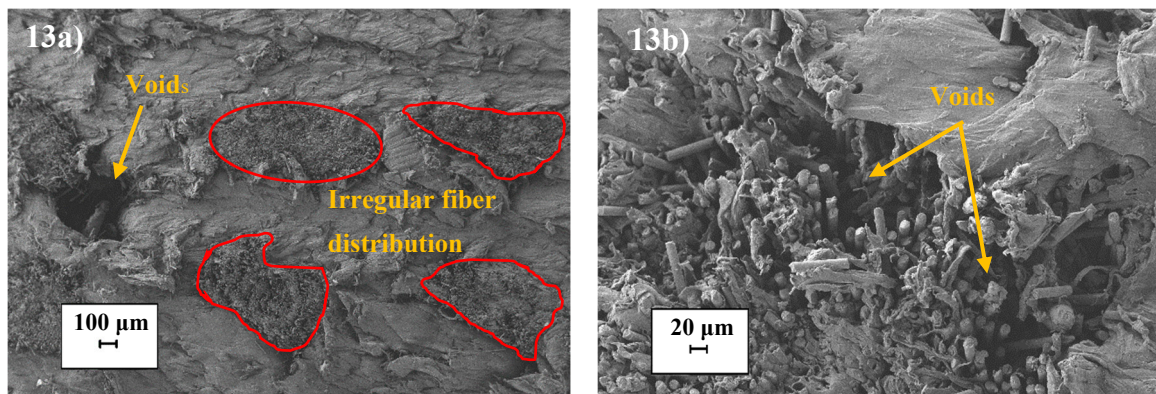
Figure 13. 3 K fibers sample before mechanical tests; (**a**) fiber distribution; (**b**) interface voids.
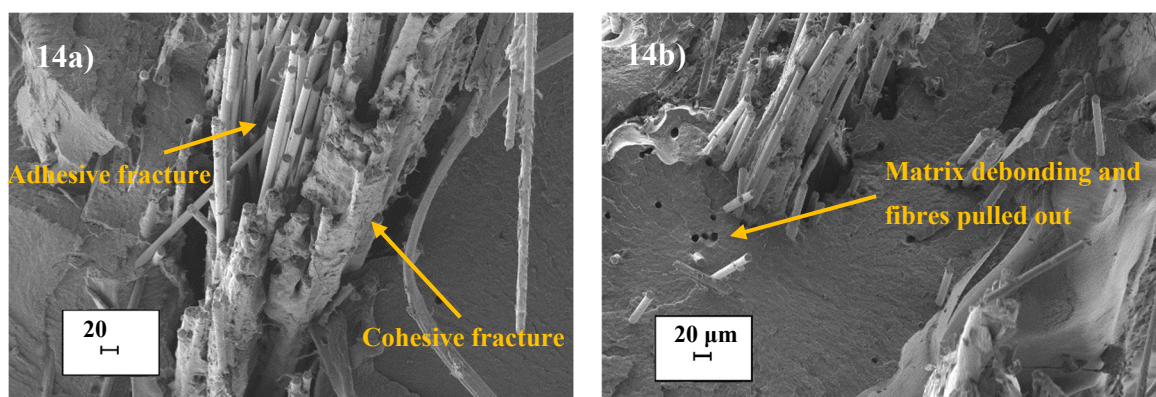

Figure 14. 3 K fibers sample after tensile tests; (**a**) matrix debonding; (**b**) carbon fibers pulled out.

The layer structure is highly visible in Figures 12a and 13a mainly due to the high temperature gradient between the previous layer and the molten polymer. Moreover, Figure 13a exhibits an irregular fiber distribution. Figure 13b shows a poor interface between polymer and fibers. Figure 14a,b shows lack of impregnation of polyamide into the fibers bundle. The voids and irregular fiber distribution can avoid the force to be transferred in a proper way, generating a matrix debonding, which finally pulls out carbon fibers (Figure 14b). Moreover, fiber–matrix interface failure (adhesive fracture), due to tensile stress concentration in the fiber–matrix interface, is observed in Figure 14a.

4. Conclusions and Future Developments

In this work, a 3D printing system was designed and built for processing (deposition of the reinforcement based on pre-defined orientations according to the commissioning of the component) and consolidation of fibers, continuous or long, pre-impregnated or not, to produce components in composite materials reinforced with carbon fibers or others by additive manufacture. The low-cost system has been developed to function in an integrated and complementary way with a common filamentary fusing additive (FFF) manufacturing equipment.

After the construction of the deposition system, its proper functioning allowed the successful printing of parts. The composite components of the polymer matrix produced have characteristics of final components with no need for post-processing operations. Additionally, and to mechanically and morphologically characterize the material processed, mechanical and SEM tests were performed on the components produced. It was concluded that the composites produced with 3 K tensile strength obtained a mean breakdown magnitude of 153.62 MPa and a mean displacement at break of 2.12 mm, and this breaking stress agrees with the low-density fraction of fibers present in the composite with anisotropic properties, resulting from the manufacturing process and the lack of optimization of the processing parameters. Regarding the mechanical behavior at bending, the composites produced

showed that with the increase of layers to double, from 2 to 4 layers, a significant increase in bending strength at about 366% and 325% for 3 K and 1 K, respectively, was obtained. The composites produced with 3 K yarns obtained double the value of the tensile strength when compared to the value obtained with 1 K yarn due to the increase of 300% in the number of fibers present in the composite. The standard deviation, both maximum force and of the breakdown voltage, reveals the need to optimize the processing parameters, namely, to improve adhesion between filaments of the same layer or between layers as demonstrated by SEM image analyses. After the beginning of the rupture, it is verified that its propagation occurs by levels, and this fact may indicate a decrease in resistance due to inter-filament rupture.

To reduce or even eliminate the problem of poor interfilamentar adhesion, a closed construction chamber must be designed and applied to the 3D printer in order to allow temperature control between the environment and the component during its construction. This effective control of the temperature of the construction envelope will allow for the study of the influence of temperature on adhesion between strands of the same layer, consecutive layers, and the percentage of voids in the components produced. The construction of a closed chamber will also enable the implementation of a filter system of the volatile organic compounds which are formed during the polymer melt process, contributing to operator safety.

The construction platform should also allow us to reach temperatures up to 150 °C to study the influence of the temperature on the adhesion of the component to the construction platform, avoiding warping, twists and detaching of the part during manufacture. The influence of the temperature of the construction platform on the mechanical properties of the produced parts, as well as the possibility of joining the fiber after the melting of the polymer filament, should also be studied. For this purpose, the metallic materials used in the construction of the heated nozzle should be a study target. Another important and pertinent future work will include the study of the possibility of using the fiber deposition system in the manufacturing of 3D composites by integrating the system into an equipment with 5 or more degrees of freedom.

The processing parameters such as temperature, deposition velocity, layer thickness and percentage of layer overlay, among others, should be the subject of future work in order to study their influence on the mechanical properties of produced parts. The optimization of the parameters of processing and maximization of the mass fraction of reinforcing fibers in the components produced should also be studied. Another interesting issue will be the pre-processing algorithm development to enable the selection of the number of reinforcement layers and to generate the production path properties file automatically from a single component file to be produced, in which the rules are already defined to overcome the limitations of the process. Lastly, other types of polymers for the composite matrix should be explored, namely the use of polyamides with lower melting points, as well as the possibility of combining the use of thermosetting resins in post-processing operations to reduce the number of voids and to increase the fibers/matrix adhesion. The influence of the variation of the diameter of the extrusion nozzle on the relationship between the fiber and the polymer should also be explored.

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