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Study of Reinforcement Distribution, Adhesion between Layers, and Porosity Induced by FDM

Arthur Wernke[1], Rafael dos Santos[2], Leonardo Santos[2], Manuel Barcelos Jr[2] and Emmanuel Lima[2]

1. Laboratório Associado de Sensores e Materiais, Instituto Nacional de Pesquisas Espaciais (INPE), Avenida dos Astronautas 1758, São José dos Campos-SP 12245-970, Brazil

2. Faculdade do Gama (FGA), Universidade de Brasília (UnB), St. Leste Projeção A, Gama-DF 72444-240, Brazil

Abstract: This study aims to understand the distribution of reinforcement material in the matrix, evaluate the adherence between layers, and determine the air gap between printing roads. We printed the specimen with two different composite materials, Polylactic Acid (PLA) reinforced with acrylic particles, and another filament reinforced with short carbon fibers. For the observations of the samples, we used a Confocal Microscope. We estimated the porosity of the material by comparing the expected mass with that achieved after manufacture. By pixel count, after binarization, we found the average percentage of acrylate particulate. They showed fair distribution through the PLA matrix even after the manufacturing process. The determination of fibers alignment was made by binarization of image, together with k-means and edge detection. This combination of methods allows estimating the fiber alignment by orientation straight lines. The manufacturing process did not offer good alignment of the fibers, even with the filament initially well aligned.

Key words: 3D printing, porosity in 3D printed parts, 3D printed composite materials.

1. Introduction

Amid the advancement of industry 4.0, emerging technologies have gained many supporters; 3D printing is one of them. Due to its versatility and low cost for rapid prototyping and manufacturing applications, there has been an exponential increase in Additive Manufacturing (AM) technology demand. This technology allows the manufacture of 3D part layer-by-layer, directly from the project without necessarily creating specific tools for each part. These advantages and the possibility to manufacture complex structures and geometries in less time or impossible to create by other processes, with micrometer resolution, helped AM become a high-demand industry [1, 2].

A standard AM method is Fused Deposition Modeling (FDM); in this method, a thermoplastic polymer's continuous filament is fed through a nozzle and heated to reach a semi-liquid state extruded on the printer bed. This process, together with plane movement, generates one printed layer, a consecutive of layers form a 3D part [2].

FDM has an advantage over other AM processes due to the facility to obtain shelf raw material. It is easier to project, share and modify the CAD part; low cost; possibility to produce a complex geometry with an office-friendly environment (simplicity), making it more popular, accessible, and flexible, but there are a limited number of materials, most commonly used with thermoplastics. On the other hand, in general, the FDM final products have limited mechanical properties compared to injected materials and a layer-by-layer appearance with low surface quality. Variation in process parameters can generate a change in manufactured parts and, consequently, in the parts' mechanical properties [3, 4]. Thus it is necessary to understand how the manufacturing process interferes with printed parts' properties to allow an evaluation to

Corresponding author: Emmanuel Lima, Ph.D., professor, research fields: phasetransformation, shape memory alloys, metallurgy and composite materials.

apply in high-level engineering projects. An option for obtaining better properties is composite materials because we can reinforce the polymeric matrix with short/long fibers, particulates, and nanoparticles that can significantly improve their properties, especially mechanical properties when reinforced with long fiber. However, the development of composite materials that meet the protocols' manufacturing requirements is necessary [5].

Particulate and short/long fibers are the most common types of reinforcements for the market's composite material. This study focuses on both of them. Particulate composite has a range of applications with a different focus, including mechanical properties improvement [6].

2. Experimental Setup

2.1 Materials and Procedures

The materials PLAC, PLAPMMA, and PLA filaments, obtained from UP3D Brasil (Valinhos, SP, Brazil), were used on 3D printer Anet A8-M, equipped with a double-extruder for polymers, classifying the process as FDM. We bought the filaments with reinforcement, and the manufacturer informed a 15% of carbon fiber reinforcement and a range of 0-30% of polymethylmethacrylate (PMMA)reinforcement; this study was determined more accurately the percentage of particulate distribution [7, 8].

Was printed the specimen by following type I of ASTM D638 for future analysis of mechanical properties. Printing parameters were 100% infill with printer roads oriented at ±45º, the temperature at 210 °C, layer height of 0.2 mm, a print speed of 50 mm/s, and a printing table temperature of 60 °C.

2.2 Test and Analysis

We analyzed the samples with OLYMPUS LEXT OLS4100 confocal laser microscope, up to 200× magnification. In PLAC filaments, the properties observed were the diameter, length, and orientation of the carbon fibers disposed within the filament. In the PLAPMMA filaments, one can observe the particulate distribution. In printed samples, we observed the same characteristics to identify variation caused by the FMD process. The pure PLA sample was used as a basis to identify the matrix in the composite material images.

We subjected the images obtained by microscopy to the binarization image processing code developed by the research group using the MATLAB software. It is useful in the characterization process because it simplifies regions of an image in black and white [9], highlighting the reinforcement of matrix and obtaining distribution information.

The process to identify the fibers' alignment was through binarization of the image to highlight the fibers, together with the k-means method, to cluster the pixels and edge detection to delimitate fiber borders. This combination of methods allows estimating the fiber orientation by straight lines (FOESL). The other properties, such as fiber diameter and length, were obtained using the confocal microscope software.

3. Results and Discussions

3.1 Voids and Adhesion between Layers

This work shows how composite parts' manufacturing process can present failure and manufacture problems through additive manufacturing of the FDM type. The evaluations of objects printed indicate a degree of porosity and irregularities.

Fig. 1a shows the void regions (dark areas) of the printed part. However, to understand the formation of voids, a closer look is necessary for the manufacturing process. The print head movement, generating spaces between the printing roads, causes the occurrence of voids. The material flow is constant during printing, but the print head is accelerated, creating irregularities in the cord width [2].

Thus the printed layer's starting point is expected to have a greater degree of voids than in the other parts of the same layer due to the print head's acceleration

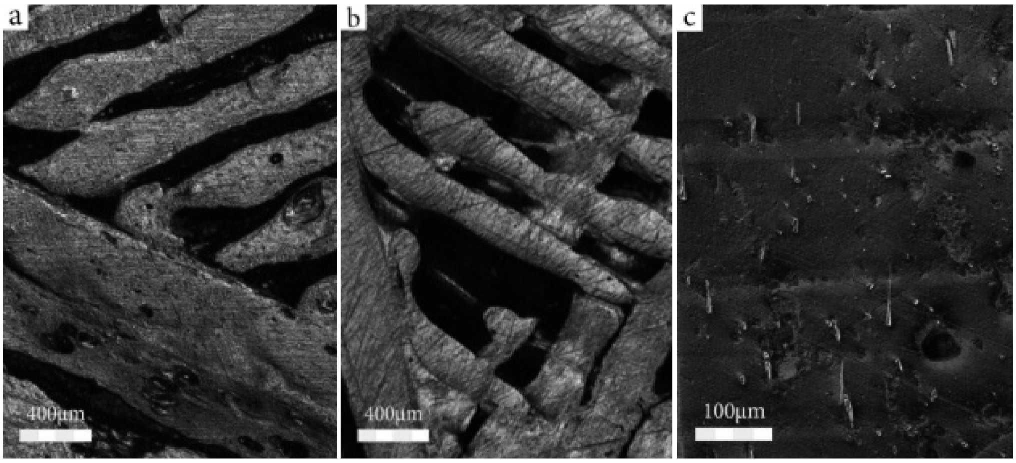

Fig. 1 Voids and gaps from the FDM process, such that (a) represents void zones (dark parts) in the middle of the piece, (b) indicates the starting point of the printed layer, the place with the highest void concentration, and (c) shows the gaps between print layers (grooves between flat areas).

and deceleration. One can corroborate this idea by Fig. 1b that demonstrates the overlap of printing voids in this region.

Fig. 1c shows the gaps (grooves between flat areas) between layers. These demonstrate the lack of adhesion between layers; the presence of this defect results in a significant reduction of mechanical properties in the direction of printed layers [10, 11].

Zhang and Wang [12] demonstrate by experimental observations and self-consistent field theory (SCFT) calculations that one can mainly attribute the layer adhesion to the large width of wetted interface and degree of inter-molecular diffusion characteristics that are influenced by the cooling rate, in consequence, influenced by printing parameters.

These showed an average porosity of 11.50 ± 0.84% for parts with acrylate reinforcement, 15.73 ± 2.33% for parts with fibrous reinforcement, and 12.20 ± 1.23% for pure PLA parts. The degree of porosity affects the mechanical properties of the part [10, 13].

The voids that occurred during the manufacturing process of printed parts by FDM alter mechanical properties, and as they appear widely distributed, hide improvements of composite materials. The printing parameters affect the specimen porosity level and, consequently, its mechanical properties [13].

3.2 Reinforcement Characteristics

Figs. 2a and 2b present the material with particulate reinforcement and its binarization zone, respectively. The binarization can estimate the average global amount of particulate reinforcement, such as 4.45 ± 0.15%. Regionally (area with 2 µm²), there is a percentage of 5.70 to 0.34%. There is a fair distribution of the reinforcing material in the polymer matrix; one can note these averages repeated in different zones and layers.

As shown in Fig. 2b, the binarization works well for highlighting the particles due to the color difference between matrix and reinforcement when observed through a microscope, allowing automatic identification via code of the percentage of reinforcement.

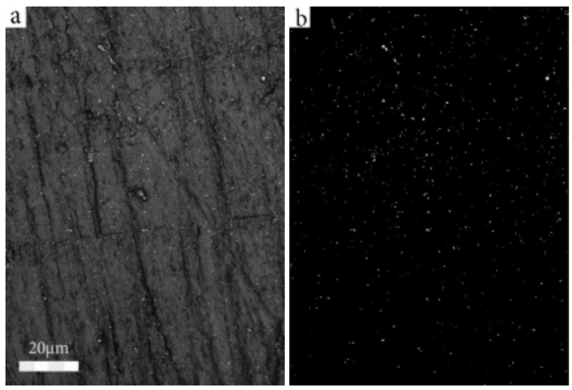

Fig. 2 Printed particulate composite, with (a) original microscopy and (b) corresponding binarized image.

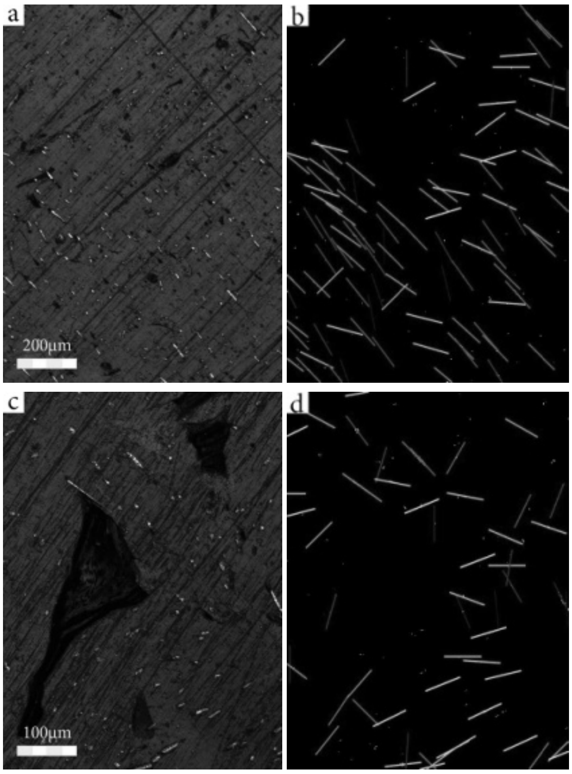

Fig. 3 Microscopy of short fiber reinforced PLA, with (a) overview of fiber alignment, (b) shows a binarized image with FOESL, (c) reports the alignment of fibers close to the microvoid, and (d) shows FOESL near the microvoid.

Fig. 3a shows the fibers' orientation, and the FOESL showed in Fig. 3b estimates the majority of the percentage lines in approximate directions, 61.3% of the fibers have their direction estimated within the main range of variation of 26.6°.

The difference in porosity between materials with different types of reinforcement, but with the same manufacturing parameters is related to extruding the fiber together from the polymer, but not controlling the pressure in the extruder nozzle. As a result, there are clogging, variations in flow, and microvoids; Fig. 3c shows the latter. This effect occurs because, during the material extrusion, no alignment or many fibers are aligned and pressed to pass through the nozzle simultaneously; therefore, there is a brief failure [1, 11].

The interruption of the extrusion flow causes a local variation of the alignment of the fibers. Fig. 3d (FOESL of Fig. 3c) presents a reduction in the fibers' alignment, totaling 25.0% from 0 to 28.2° and 16.1% from 0 to -24.8°. There is still a difference of 31.2% in the fibers' alignment, even considering both bands' junction.

Gupta et al.[14] find the tensile strength of reinforced PC is marginally less than pure PC in all three reinforcement concentrations tested (3, 5, and 7.5% by volume). They estimated, without post-processed images, that effect is caused by non-alignment in printing direction or to the loading direction, causing a stress concentration, which eventually reduces the tensile strength of the 3D composite [14]. However, Ferreira et al. [15] have found better alignment and better mechanical properties. The results presented in this study and the bibliographic review are a strong indicator of their conclusion that this alignment is a function of the production process and possibly reinforcement geometry.

Fig. 4a shows adhesion between matrix and fiber, and it is possible to notice dark areas around the fibers, which is indicative of the bad adhesion. This effect is characteristic of a PLA matrix with no treated carbon fiber reinforcement, even with pressure control in the extruding head [3]. With the adhesion's zoom observation, the fibers' non-alignment is visible, as seen in Fig. 4b, even in continuous areas, without printing voids of microvoids.

Gupta et al. [14] found adhesion problems with using polycarbonate (PC) reinforced with carbon fiber. It is a significant result to highlight FDM composite problem with adhesion between matrix and fiber.

Through measurements with a confocal microscope, it was possible to determine the main dimensions of fibers, with an average length and diameter of 149.96 μm and 6.25 μm, respectively.

3.3 Reinforcement before 3D Printing

After visualizing how reinforcing material was after printing on the specimens, it is useful to understand how the material was before the process and how the

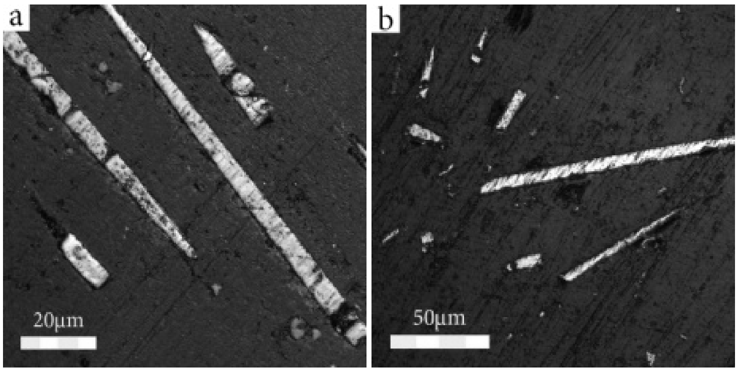

Fig. 4 Zoom on fibers for visualization of (a) the probable lack of adhesion between matrix and fiber, with spaces (dark areas) between them and (b) fibers in the zoom, reveal that, in reality, there is no alignment of the fibers.

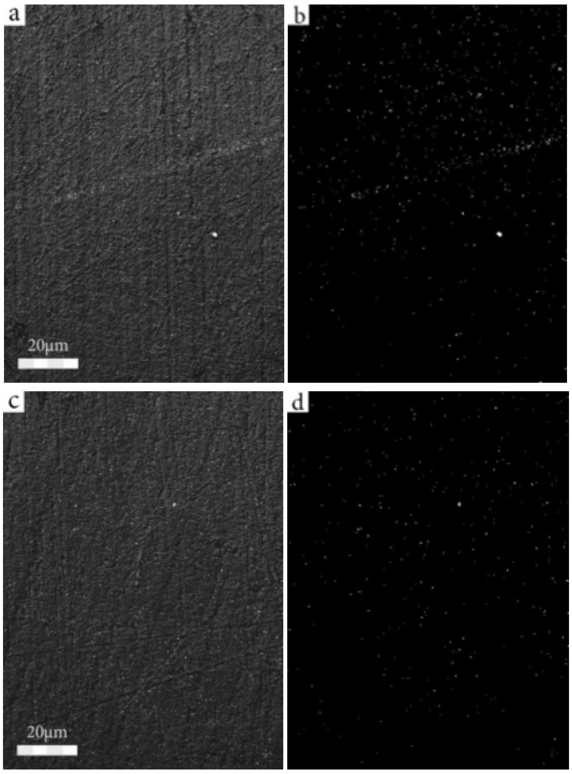

Fig. 5 Microscopy and binarization of PLA filament with PMMA, with (a) showing the cross-section of the filament, (b) its binarization, (c) showing the superficial longitudinal section of the filament, and (d) its respective binarization.

FDM process changed the composite material. PLA filament with reinforcement of PMMA has 4.30 ± 0.24% of acrylate concentration, very close to the concentration found after printing the specimens, however, locally, it presented a great variation of this percentage (of 7.53 to 0.01%), this effect is visible on the cross-section of filament, shown in Fig. 5b (binarization of Fig. 5a).

The filament has the worst quality of particulate distribution; empty zones do not happen in printed pieces; we can attribute this effect to the nozzle's material concentration. During printing, the pressure increases and deposits the melted material more homogeneously than in the original filament.

This point is evident when looking at the filament walls (Figs. 5c and 5d); they present a significant difference in the distribution of the material. However, in the printed piece, this difference has no influence. The reason is when melting the filament and passing to the extruding head, the cross-sectional area reduces, and the pressure increase results in the polymer's mixed flow.

The printed part has an equivalent distribution and in isolated places, even better than the original filament. Therefore, this study shows that the particulate composite has good compatibility with the FDM process.

The filament with short fiber reinforcement has its surface in Fig. 6a, and one can notice that the most fibers align longitudinally with the filament, which is more evident when observing the cross-section of the filament in Fig. 6c. We observed just a few fibers transversal to the main direction in any of the cuts analyzed.

When comparing the presented results between Figs. 6b and 3b, one can demonstrate, the standard FDM process used by 3D printers cannot offer good alignment of fibers, even if the initial raw material

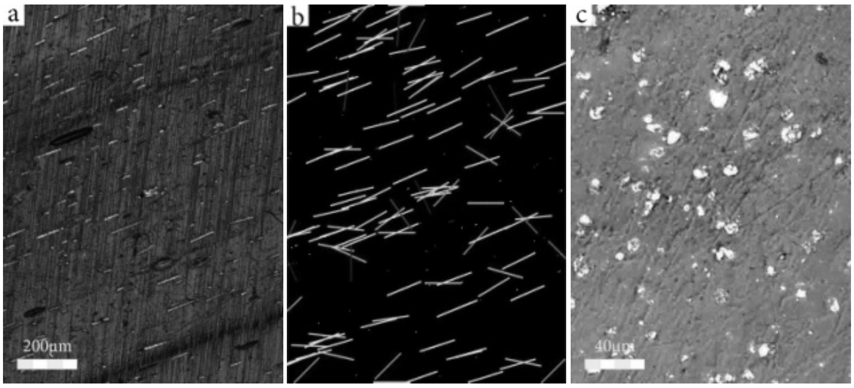

Fig. 6 Alignment of short carbon fibers in PLA filament: (a) represents the longitudinal surface of the filament, (b) shows FOESL, and (c) represents filament cross-section.

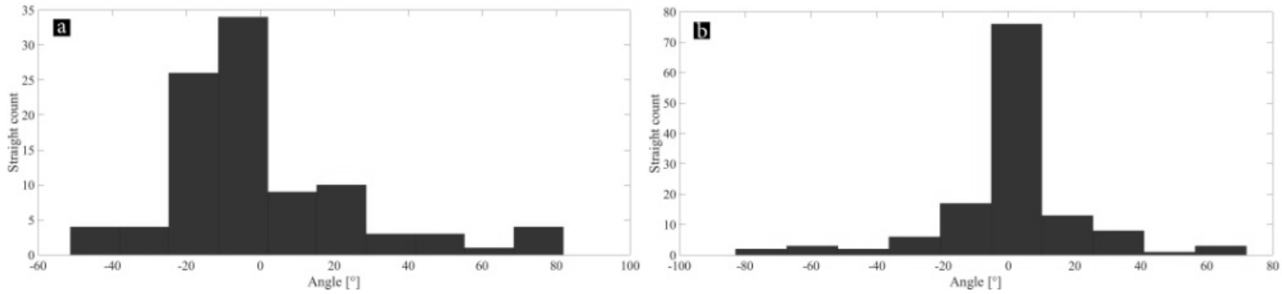

Fig. 7 Histograms refer to the amount of fiber present in each angle range, with (a) representing the straight-line count in the printed specimen and (b) representing the straight-line count in the filament before printing.

(filament) has the fibers well aligned. This effect is evident when comparing the histograms of FOESL of the printed material (Fig. 7a) with that of the filament (Fig. 7b). The distribution for printed material demonstrates the percentage of alignment already presented.

In filament distribution, it has a prominent alignment peak, and its main lines are in the range of -13.1 to 17.8°, which corresponds to 86.6% of the lines and does not present points of failure equivalent to microvoids presented in Fig. 3c.

Morrill et al. [16] used probability density function (PDF) with perfect semicircular von Mises distribution to show the relation between histogram distribution and fiber alignment. If the fibers are entirely non-aligned, the distribution is closer to uniformity, but if the fibers are well-aligned, the distribution has a concentrated peak at the primary fiber angle. However, it is possible to notice in Fig. 7, and the adjacent bands are more evident in the printed material; this indicates that, even with alignment percentage close to the filament, there is an optimum distribution between the angles bands, with more significant influence.

4. Conclusions

This work shows how composite parts' manufacturing process can present failure and manufacture problems through additive manufacturing of the FDM type. The evaluations of objects printed indicate a degree of porosity and irregularities.

Regarding the distribution of the reinforcement material in the polymer matrix, the present study showed, the pieces printed from the PLA reinforced with particulate material present an equivalent distribution in isolated locations even better than the original filament. It is worth mentioning the applicability of the pixel binarization method for this analysis, which worked well to identify the particles by image processing code automatically. On the other hand, in parts manufactured by FDM with composites reinforced with short fibers, the fibers' worst alignment is even if the filament has the fibers well aligned. Therefore, the FDM process with short fibers could not promote a good fiber alignment, at least without advanced controls of pressure in extruding head and polymer flow.

This study presented aspects of composite parts' production using 3D printing, showing some problems. However, identifying these anomalies adds knowledge for improved AM technology in composite materials, especially in projects that demand greater rigor. Future research needs to understand which parameter produces better fibers' alignment and adhesion between matrix and fiber and understand how particulate and fiber concentration influence composite material optimization.

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