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Recibido: 26-11-2022 Aceptado:15 -05-2023 Disponible online: 01-06-2023 Angle influence and Toughness characterization of bioinspired discontinuous fiber helicoids composite materials produced via additive manufacturing

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Abstract

Nature has evolved composite materials over millennia that are compatible between stiffness and toughness and that exhibit outstanding mechanical characteristics compared to the low properties of their base materials; as a result, they are an excellent source of inspiration for material optimization for applications such as increasing toughness and damage resistance, an aspect that is challenging in conventional engineering materials. Nowadays eight structural elements are identified in biological materials: fibrous, helicoidal, gradients, layered, tubular, cellular, suture, and overlapping. Helical structures consist of stacks of ordered fibers that form layers that are rotated at a constant angle of inclination. These include plywood and Bouligand structures. Bouligand structures consist of an arrangement of fibrous laminates that completes a 180° turn and provides some biological materials with enhanced strength and toughness in multiple directions and exceptionally high fracture resistance. The classical composite materials mechanics provides some constitutive model approximations for this type of materials, but they still need to be studied and tested to properly understand their behavior. Injection molding, compression molding, hand layup, resin transfer molding, filament winding, pultrusion, and automated fiber placement are just a few of the traditional methods used to make fiber-reinforced polymer composites (FRPC). However, these traditional manufacturing techniques have a restriction on specific fiber alignment and demand expensive molds, dies, or lithographic masks. Additive manufacturing has the potential to replace many conventional manufacturing processes due to its ability to create complex geometries with customizable material properties and employ several materials simultaneously, among other things. Fused deposition modeling (FDM) is the most widely used manufacturing additive technique for manufacturing FRPC due to its low cost, low energy input, material consumption, and operation simplicity. This work presents three-dimensional models mimicking the Bouligand structures by turning the pitch angle of the layers and an analytical model comparison were made. The specimens were fabricated using the FDM technique. Thermoplastic polyurethane (TPU) was used for the matrix and polylactic acid (PLA) for the fibers. Tensile tests were performed to determine the impact of the helicoidal angle and the matrix and fiber materials on the stiffness, strength, and toughness of the laminated composite. To determine the effect of the helical angle and the contribution of the matrix and fiber components on the strength and toughness of the laminated composite, tensile tests were performed. Analysis and experimentation suggest that increasing the pitch angle enhances the composite's strength, stiffness, and toughness.

Key words: Biomimetic, Bouligand structures, Fiber reinforced polymer composite, Additive manufacturing, Stiffness, Toughness.

Influencia del ángulo y caracterización de la tenacidad en materiales compuestos bioinspirados con fibras helicoidales discontinuas fabricados con manufactura aditiva

1. Introduction

In the research to create materials with improved mechanical performance, biomimetics plays a very important role today. The result of the observation and imitation of the solutions that nature employs throughout the evolutionary process can contribute to areas such as the creation of protective structures, aerodynamic elements, weight reduction, thermal transfer, and optimization of materials, among others. Bio-inspired structures have been employed in a wide range of applications such as in the design of automobiles and trains, for building structures, aircraft wings, missile structures, protective armor, and unmanned aerial vehicle (UAV) design, among others (Hu et al., 2019; Wang et al., 2020)(Hu et al., 2019; Wang et al., 2020). It has been established that hierarchical structures provide an adequate distribution, dimensions, and properties to the components of a system so that it responds optimally to a particular need of a species(Ikoma *et al.*, 2003). There are a variety of hierarchical structures; some are comprised of fibers, plates, particles or inclusions, pores, multilayer configurations, organic and inorganic interfaces. According to the organization the fiber structures can be categorized as unidirectional, orthogonal, and helical (Zelazny and Neville, 1972; Wang et al., 2020) using Oryctes rhinoceros and five

other species of beetles. The precision of orientation of each specific endocuticle layer at a given location is shown by the low variation $(\pm 3.3^{\circ} \text{ for 10} \text{ measurements}$. Fiber reinforced composites (FRC) are the materials that structurally resemble bio-inspired structures the most since they can be manufactured to have better properties than the sum of their parts [5][6], displaying an anisotropic and heterogeneous behavior that is different from that of conventional materials (Arias and Vanegas, 2004) (Yang, Yi and Xie, 2021) (Li *et al.*, 2022) (Liu *et al.*, 2022). On the other hand, Bouligand structures consist of an arrangement of fibrous laminates that completes a 180° turn and provides some biological materials with strength and toughness in multiple directions and exceptionally high fracture toughness. The properties of a composite material depend on the properties of the constituent materials, their geometry, distribution, and volumetric fraction.

The bio-inspired composite material can be analyzed as a laminae-type composite, where the thickness of a layer is orders of magnitude less than the other dimensions of the layer. Therefore, they have been analyzed as plate-like elements. Where the constitutive equation for FRC with fibers oriented at 0° is shown in equation (1) (Reddy, 1999).

$$\{\epsilon\} = [s] \cdot \{\sigma\} \tag{1}$$

Where, $\{\epsilon\}$ is strain vector, [s] is compliance matrix, and $\{\sigma\}$ is stress vector. This expression can also be given in terms of its global components x and y, when fibers are not aligned with the loading direction. The engineering constants of the non-oriented lamina can be determined directly from the elastic constants and the orientation angle (see equations (2) and (3)).

$$\{\varepsilon_{xy}\} = \overline{[S]} \cdot \{\sigma_{xy}\}$$
(2)

$$S = [T_{\varepsilon}(\theta)^{-1} \cdot \overline{[S]} \cdot [T_{\sigma}(\theta)]$$
(3)

Where: $\{\varepsilon_{xy}\}, \{\sigma_{xy}\}$ correspond to the normal strains and x-y stresses, respectively, $\overline{[S]}$ is global compliance matrix and $[T_{\epsilon}(\theta)], [T_{\sigma}(\theta)]$ are the coordinate transformation matrices for the deformation and stress vectors. From the above equations it follows the equations for laminated materials.

$$\{N M\} = [A B B D] \cdot \{\varepsilon k\}$$
(4)

Inside this equation the design variables for thin laminates are involved, the forces in the plane {N} and the moments {M} are related with strain { ϵ } and bending {k} (Arias and Vanegas, 2004; Kienzler, Ott and Altenbach, 2004).

Method Equations			
	$P/P_{m} = (1 + \xi \mu V_{f})/(1 - \mu V_{f})$		
Rule of mixtures (ROM) (Huang <i>et al.</i> , 2021)	$\mu = (P_f/P_m - 1)/(P_f/P_m + 1)$	(5)	
($E_1 = E_f V_f + E_m V_m$		
	$E_1 = \eta V_f E_f + (1 - V_f) E_m$		
Shear-lag (Shokrieh and	$\eta=1-tanh(\beta l/2)/(\beta l/2)$		
Moshrefzadeh-Sani, 2016;	$\beta = \sqrt{(I/(\pi a^2 E_f))}$	(6)	
Huang <i>et al.</i> , 2021)	$I=(2\pi G_m)/\ln(b/a)$		
	$b/a = \sqrt{(2\pi/(\sqrt{3} V_f))}$		
Halpin-Tsai (Luo <i>et al.,</i>	$E_{1}/E_{m} = (1 + \xi \eta V_{f})/(1 - \eta V_{f})$		
2018; Huang et al., 2021)	$\eta = (E_f/E_m - 1)/(E_f/E_m + 1)$	(7)	

In comparison to continuous fiber composites, short fiber reinforced composites have superior machinability, reduced manufacturing costs, and more automated production. Young's modulus of these composites depends on the elastic properties of the matrix and the fibers, volume fraction and the aspect ratio of the fibers. There are several methods for determining the elastic properties of short fiber reinforced composites. In this study, we will concentrate on three of the methods for measuring the elastic characteristics of short fiber reinforced composites (Table 1).

Due to its many advantages and affordable prices, additive manufacturing is currently a widely used manufacturing process. Additionally, the technology allows the development of complex structures with adequate dimensional control and the use of multiple materials, even at the same time. All of these attributes make additive manufacturing an appropriate tool in the research process for the optimization of materials in engineering [14] as well as for the production of models and some finished products (Gao *et al.*, 2015; Huang *et al.*, 2015; Carvajal Loaiza *et al.*, 2020).

This article is an extension of the paper originally presented in EXPOIngeniería 2022 ('Engineering for ENERGY INFRASTRUCTURE 4.0 TECHNOLOGY VITAL ENGINEERING Facultad de Ingeniería', no date), we combine two architectures: the helicoidal or Bouligand (Chen et al., 2008; Meyers and Hodge, 2008) and the "brick and mortar" (Barthelat et al., 2007; Meyers et al., 2008; Wang et al., 2020) to investigate, discontinuous fiber helicoids (DFH) composites as function of helicoidal angle between layers. The fiber helicoid is found in the dactyl club of the Peacock mantis shrimp (Stomatopod) (Weaver et al., 2012; Grunenfelder et al., 2014; Guarín-Zapata et al., 2015), the cuticle of arthropods (Cheng, Wang and Karlsson, 2008; Cheng et al., 2011; Weaver et al., 2012; Grunenfelder et al., 2014; Chen et al., 2018) and fish scales (Ikoma et al., 2003; Bruet et al., 2008; Torres et al., 2008; Chen et al., 2012; Zhu, Ortega, et al., 2012; Zhu, Szewciw, et al., 2012; Zimmermann et al., 2013; Fang et al., 2014; Gil-Duran, Arola and Ossa, 2015; Murcia et al., 2016; Yang et al., 2019). The brick and mortar is found in sea shells, e.g., the innermost layers of red abalone (Haliotis rufescens) (Ji and Gao, 2004; Salinas and Kisailus, 2013; Barthelat, Yin and Buehler, 2016). It has been proposed that helicoidal arrays of fibrils, present in Arapaima Gigas scales, adapt to the loading environment through laminae rotation towards the loading direction, whereas other laminae, with large offaxis angle, rotate away from the loading direction(Zimmermann et al., 2013; Yang et al., 2014; Suksangpanya et al., 2018). Also, this fiber reorientation has been found to contribute to enhanced ductility and toughness in fish scales (Yang et al., 2019). For this reason, here, we fabricated a discontinuous Bouligand assembly composed of fibers of PLA in embedded in a TPU matrix to investigate the effect of the angle between layers on the strength, stiffness, and toughness of the helicoidal composite.

2. Methodology

The principal objective of this research is to increase the material toughness, and as a result, a Bouligand-type arrangement was chosen. This type of configuration contributing to the toughness and ductility of the armor or shell by layers reorienting towards the axis of stress and deforming by stretching, sliding, and delamination mechanisms while other layers rotate away from the tension and compression axis.

The Fused deposition modeling (FDM) technique was utilized to fabricate the specimens with a Bouligand arrangement, due to its low cost, the possibility of printing two materials simultaneously. simplicity of usage, and variety of material options. Considering that the matrix must be a soft material compared to the fibers, the material used for the matrix was thermoplastic polyurethane (TPU) and polylactic acid (PLA) for the fibers. These materials provide a difference of about two orders of magnitude in their Young's modulus with which it is possible to simulate the materials used by nature. The discontinuous fiber helicoid (DFH) composites are 3D printed with two materials. The fibers were printed with a square cross-section area. The stiff fibers have a length of 10 mm, a square cross-section with a width of 1.2 mm, and an aspect ratio of the gauge, where W and L were the width and length of the specimen respectively. The matrix wraps the fibers in all directions by a thickness of 1.2 mm. The matrix thickness is a function of the number of necessary layers to complete a Bouligand structure unit (180°), see figure 1.

Figure 1. A) Examples of animals with Bouligand type arrangement, B) Bouligand type arrangement C) CAD sectional view of a Bouligand type test specimen with discontinuous fibers for a pitch angle of 45° and D) example of a specimen assembly in the test machine. Source: authors ownership.



Inventor 2019 Software(Autodesk, no date) was used to create the design and CAD models of the specimens, which were then exported in STL format to Cura 4.6 (Ultimaker, no date), an open-source program, where G codes were generated. An Ultimaker S5 printer was used for manufacturing (Ultimaker, no date), with an extrusion temperature of 225 °C, 100 % as infill percentage, and the layer height was 0.04 mm, for both materials, and extruder speeds were 25 mm/s for TPU and 70 mm/s for PLA.

Mechanical tests were performed at the materials laboratory of the Universidad de Antioquia. The printed composites were tested under guasi-static tensile loading at a strain rate of $2.77 \times 10^{-4} \text{ s}^{-1}$ inside a universal testing machine, Shimadzu AGX Plus equipped with a 10kN load cell. The mechanical characterization of the bulk materials was carried out according to ASTM D638 Standard *Test Method for Tensile Properties of Plastics* and an Epsilon axial extensometer 3542-050M-020-ST was used to determine the elastic modulus of TPU and PLA. For all tests, the loading direction was parallel to fibers of the first layer printed in the helicoid. Fiber orientation in subsequent layers was defined by the pitch angle of each specimen. The studied pitch angles were 30°, 45° and 60° and a set of five tests were made for each one and compare with analytical Shear-lag model, Halpin-Tsai's formulae, and rule of mixtures. The incidence of pitch angle on the stiffness, strength and toughness of composites was compared and analyzed. The toughness was determined as the area under the strain stress curve.

3. Results and discussion

Materials used for the matrix and fibers were characterized, obtained values can be observed in table 2 and their mechanical behavior can be seen in

Tak	ole 2. Mechanical properti	properties of materials used to manufacture the specimens. Source: authors ownership.				
	Material	Young's modulus [MPa]	Poisson's ratio			
	TPU (matrix)	16.67	0.45			
	PLA (fibers)	1303.4	0.37			

The stress-strain relationships obtained for the angles of 30° , 45° , and 60° can be seen in figure 2. These angles were measured from loading axis. For all the tested specimens, two zones were observed, zone 1 with a close to linear behavior where the contribution in the resistance is given by the fibers and zone 2 with a plastic behavior with a little strain hardening due to the realignment of the fibers. This result has already been observed and discussed by other authors (Wang *et al.*, 2020; Wu *et al.*, 2020).

The strength of the test specimen in contrast to the bulk material used as the matrix and fibers is shown in figure 3. A combination of the matrix and fiber strengths was obtained, and it was observed that the resistance of this Bouligand structural composite material fabricated via additive manufacturing complies with the mechanics of materials (Arias and Vanegas, 2004; Pérez and Sánchez, 2014). For the angles analyzed, the pitch angle with the highest toughness was 45° and the one with the lowest toughness was 60°. Currently, there are many differences among researchers regarding the optimal pitch angle to improve energy absorption. Wenting et al, Sha Yin et al., Kaijin Wu et al., Yuan et al, and Jiahua et al., (Wu et al., 2020; Yin et al., 2020; Ni et al., 2021; Ouyang et al., 2021; Chen et al., 2022; Yuan et al., 2022), reported optimum pitch angle values of 9.1°, 16°, 20°, 25°, 30° and 36° respectively. These variations could result from a variety of factors, including the materials employed, the composite scale (nano, micro and macro) and the sorts of testing carried out. The type of tests performed includes fracture mechanics (Wu et al., 2020; Körbelin et al., 2021; Chen et al., 2022), impact resistance (Yuan et al., 2022), and tensile strength (Huang *et al.*, 2021).

Stiffness, tensile strength, and toughness according to pitch angle can be seen in figure 4. This graph shows the influence of the fibers angle on the behavior at zone 1 of a DFH material. An increase in stiffness of composite can be observed as the angle increases while the highest value for the strength was obtained for an angle of 45°. This can be explained by the fact that this angle is the one that evenly distributes the properties in both the direction of the applied load and perpendicular to it. Similar findings were reported by (Yin *et al.*, 2020) for most of their tests. In specimens with a helical structure printed using the same FDM technology but with a polyamide and polypropylene matrix, the pitch angle was adjusted from 10° to 45°. Additionally, it was noted that the fluctuation of the plateau stress decreased as the angle increased.





Toughness is a mechanical property that is crucial when designing materials that must exhibit excellent penetration resistance and fracture toughness characteristics. Nature combines various mechanical structures, phase organization, and constituent selections to achieve these properties. These materials could be employed for biological applications, wearable technology, and protection structures, among other things. In this study, toughness showed comparable values in all cases, but the greatest average values were reached by 45° pitch angle, and these values increased by around 10%. When the pitch angle between layers increases, the soft matrix material contributes more, which causes this increase. This finding may help designers develop materials with enhaced mechanical properties.



For analytical calculations $V_f = 0.462$ and $V_m = 0.538$, these values were calculated by CAD models, the results for the calculation of Composite Young's Modulus for each method is sow in table 3.



Table 3: Composite Young's Modulus for each method analyzed. Source: authors ownership.						
Fiber angle		30°	45°	60°		
Composite Young's Modulus	ROM Shear-lag Halpin-Tsai	102.653 22.305 60.144	128.700 22.086 69.906	151.254 21.918 76.803		

Elastic property of zone 1 is highly method-dependent, ROM and Halpin-Tsai method shows an increase in Young's modulus as the fiber angle increases, while for the Shear-lag method it remains almost constant. Compared to the experimental values (figure 4), the ROM method is the most accurate and simplest one. When the stresses acting on the part involve mainly shear, it is most appropriate to use the other methods for estimating the elastic properties of short fiber composites.

4. Conclusions

A helpful manufacturing technique for testing and research on twomaterial composites is additive manufacturing. Mechanical tests of these printed specimens yield important data for the design of bioinspired materials.

The mechanical properties of composite materials manufactured via additive manufacturing are significantly influenced by the pitch angle between layers. By incorporating discontinuous fibers in a composite with a Bouligand structure, a composite material with two remarkable defined deformation zones was obtained. A plastic zone where the realignment and geometric rearrangement of the fibers produce a strain hardening effect and a nearly linear zone where the fibers offer resistance. As a result, for custom - built material, it is possible to improve toughness, stiffness, and strength.

As the pitch angle increased, an increase in the analytical and experimental resistance of composite was obtained. The fiber contribution is stronger for smaller pitch angle values. This result agrees with what is established in the mechanics of composite materials.

The 45° pitch angle, which also showed an increase in plateau stress and an improvement in resistance compared to fibers orientated at 30°, demonstrated an enhanced mechanical behavior when increasing the pitch angle of fibers. It is advised to

utilize a helicoidal angle of 45 degrees for applications requiring outstanding toughness.

Short-fiber composite behavior is generally not predictable by analytical or empirical methods. In order to validate the behavior of these materials, simulations, and experiments in laboratories need to be conducted. However, it is important to keep in mind that the final shape of the component might also influence the composite's behavior.

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