

**Mechanical properties characterization of specimens
made by additive manufacturing reinforced with
natural fibers**

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O sucesso nasce do querer, da determinação e persistência em se chegar a um objetivo.

(José de Alencar)

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Resumo

Face á grande preocupação ambiental que tem surgido nos últimos anos, e a necessidade de substituir os polímeros provenientes do petróleo, têm sido estudados os compósitos reforçados com fibras naturais, como alternativa amiga do ambiente. Estes compósitos têm como aplicação na indústria automóvel, aeroespacial, onde é necessário fazer uma redução do peso dos componentes.

No presente trabalho, pretende-se estudar a influencia que as fibras de linho e de coco têm no aumento da resistência dos compósitos reforçados com estas, à flexão e à tração. Na literatura são sugeridas várias técnicas para melhorar as propriedades das fibras, portanto, neste estudo foi testado um tratamento químico, alcalino com NaOH, e uma técnica física, de disposição das fibras em forma de rede.

Após análise dos resultados, verificou-se que a percentagem de enchimento é a propriedade com maior influência para aumento das propriedades mecânicas estudadas. Por outro lado, verificou-se que a utilização do tratamento alcalino não fez diferença nos ensaios de tração das fibras de linho. Quando à aplicação das fibras em rede, estas obtiveram ainda piores resultados, ao contrário do esperado.

Verificou-se que as combinações ideais alteram de ensaio para ensaio.

Abstract

Given the great environmental concern that has arisen in recent years, and the need to replace petroleum-based polymers, composites reinforced with natural fibers have been studied as an environmentally friendly alternative. These composites have applications in the automotive and aerospace industries, where it is necessary to reduce the weight of the components.

In the present work, we intend to study the influence that flax and coir fibers have in increasing the flexural and tensile strength of composites reinforced with them. In the literature several techniques are suggested to improve the properties of the fibers, therefore, in this study a chemical treatment, alkaline with NaOH, and a physical technique, of arranging the fibers in a network form, were tested.

After analyzing the results, it was found that the percentage of filler is the property with the greatest influence for increasing the mechanical properties studied. On the other hand, it was found that the use of the alkaline treatment made no difference in the tensile tests of the flax fibers. When it came to the application of the networked fibers, they obtained even worse results, contrary to what was expected.

It has been found that the ideal combinations change from trial to trial.

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List of Abbreviations and Symbols

Abbreviations

3D	Three dimensions
Adj MS	Mean squares
Adj SS	Sum of squares
DF	Degree of Freedom
HG	Homogalacturonan
MDF	Medium Density Fiberboard
NaOH	Sodium Hydroxide
NFPCs	Natural fiber reinforced polymer composites
NFRCs	Natural fiber reinforced composites
OH	Hydroxyl
PBS	Polybutylene succinate
PHB	Polyhydroxybutyrate
PLA	Polylactic Acid
S/N	Signal-to-Noise
UV	Ultraviolet

Symbols:

A	Mean cross-sectional area	mm ²
b	Specimen width	mm
h	Thickness	mm
L	Specimen length	mm
L ₀	Initial length of specimen	mm
P	Load applied at the point	N
V	Test speed	mm/min
ε	Strain	-
ε'	Deformation rate of 1%/min	-
σ	Axial tensile stress at one point	MPa

Chapter 1

Introduction

1.1 Framing

Nowadays, natural and synthetic fibers are increasingly in demand for various industrial applications and are used in the automotive and aerospace sectors to reduce the weight of materials. Natural fibers have immense advantages being able to stand out the low cost, abundant availability, ability to control the ecosystem of the environment, biodegradability and sustainability, as well as their low density and high specific characteristics [1].

The role of fibers in fiber-reinforced composites is essentially to adjust the characteristics of the materials to achieve increased durability or flexibility. Natural and synthetic fibers are classified according to their origin [2,3].

Natural fibers are divided into three groups, animal, mineral, and vegetable. Animal fibers are, for example, silk, which comes from the hair of goats, sheep, and horses, and wool, which comes from the wool of sheep, alpaca, and camel, among others. Mineral fibers are those that come from asbestos, glass, and graphite. Plant fibers, on the other hand, are the most accepted by industry to replace synthetic chemicals and non-renewable resources. These can be wood, grass, fruit e.g. coconut, fibrous material derived from hemp, knaf, flax and jute, and leaves from banana and cotton [4].

Flax fibers are considered one of the best among natural fibers in terms of tensile strength and abundance. Numerous researchers have been conducted to improve the interphase adhesion of flax fibers with a polymeric matrix, the alkaline treatment is the simplest and most efficient treatment [5].

Coconut fiber is extracted from the outer shell of the coconut. The common name, scientific name and plant family of coconut fiber is Coir, *Cocos nucifera* and Arecaceae (Palm), respectively. This fiber is one of the most abundant fibers in tropical countries such as in India, Sri Lanka, Thailand, and Brazil, among others. Coconut fibers have many advantages, they are moth proof, resistant to fungus and rot, have good insulation against temperature and sound, are not easily combustible, flame retardant, unaffected by moisture, strong and durable, and return to shape even after constant use [6].

There are several methods to improve the mechanical and physical properties of natural fibers, and this is an active topic that is being studied and developed day by day. You can achieve a better performance in one area by being quite different in another, and not result in the same way. For example, increasing the stiffness of a material may be advantageous in construction, while in biological applications improvements in toughness or flexibility may be preferred [7].

There are several techniques to change the fiber strength, including physical and chemical techniques. The physical technique has been the most widely used for several centuries for altering fibers for various purposes, such as ropes, clothing and baskets, using the physical procedures of bundling plant fibers separated into distinct threads [4].

Regarding chemical techniques, this method uses chemical reagents to alter the surface properties of natural fibers with the intention of removing the poorer end of the natural fiber layers, changing the alkalinity and acidity of the fibers, and generating a consistent, flexible layer on top of the natural fiber layers [8]. There is a treatment of natural fibers to alter the chemical composition of the cellulosic polymer, which is an alkaline treatment using sodium hydroxide (NaOH) [4]. This treatment makes the fiber clearer and more uniform, that is, the fiber surface develops more evenly as micro voids are eliminated, resulting in the strengthening of the pressure displacement capacity between the end cells. In addition, the alkaline treatment decreases the fiber diameter, increasing the length [9]. With this treatment the mechanical and thermal characteristics of the composite are greatly improved [10,11].

Synthetic fibers can have a polymeric or mineral-based composition, some examples being fiberglass and carbon. Depending on their properties, they can be elastic, flexible, rigid, and can have lower or higher water absorption. Compared to natural fibers, they are more resistant to water, stains, heat, and chemical damage. In general, natural fibers are easier to break down chemically than synthetic fibers, because they are biodegradable and can be attacked by bacteria and fungi. In opposition, most synthetic fibers are not biodegradable and wear out over time, while natural fibers can be easily broken down [12].

Although the use of natural fibers partially satisfies the requirements of regulations that mandate the use of environmentally friendly and sustainable materials. The consumption of petroleum-based plastics has grown more and more, and the

depletion of this raw material has led to the search for materials from natural sources, an example being lignocellulosic waste. So, the development of these completely biodegradable materials like polymers (polylactic acid (PLA)) have generated a great interest [13–17]. PLA is one of the best biopolymers because of its characteristics; it has properties such as elasticity, stiffness, thermoplastic behavior, biocompatibility, and good moldability. It is produced from lactic acid, which in turn can be produced biotechnologically using renewable natural resources such as corn and sugarcane starch [18,19]. By adding additives such as metals, metal oxides, antibiotics, enzymes and natural compounds, this polymer can be given several functions [19,20].

3D printing is one of the revolutionary techniques in the field of manufacturing, and PLA is the most widely used filament for printing. 3D printing represents a "material by design" approach in the manufacturing process with a very high degree of freedom, as it allows functional adaptation of the mesostructure of the material, i.e. the layer-scale structure [21–23]. When designing the organization of the material through the deposition process, the structures are functionally classified to have the desired amount of material (thickness and width) [24,25].

PLA has the advantage of being biodegradable, however, this filament in pure form is brittle and has low toughness and flexibility. To overcome these disadvantages, PLA composites are used [5]. The combination of PLA with natural fibers are mainly used as low-cost materials that have functional structural properties. Because petrochemical materials are a finite resource, there is an alternative to materials based on renewable resources [26]. Natural fibers such as flax and coconut fiber are blended with polymers such as PLA, because their constituents (starch) make it possible to increase impact strength and decrease water absorption [27,28]. The tensile properties of the biocomposite improve with the addition of the natural fiber, depending on the nature of the fiber, its orientation, content, and shape (fiber or fabric) used [29].

Due to all these aspects, the objectives of this study are presented in the next subchapter.

1.2 Motivation and Goals

This dissertation reports on the study of mechanical properties characterization of specimens made by additive manufacturing reinforced with natural fibers. The main purpose of this study is to determine the mechanical properties of composites reinforced

with the natural fibers flax and coconut. In order to achieve this goal, it is necessary to go through several steps, which are as follows:

- Define what types of fibers exist and address the properties of the fibers under study;
- State the properties of the polymers (PLA);
- To study the influence of a flax fiber reinforcement in a PLA composite and determine the optimal conditions to increase the tensile and flexural strength;
- To study the influence of a coconut fiber reinforcement in a PLA composite and determine the optimal conditions to increase tensile and flexural strength;
- Make a comparison between composites reinforced with flax fiber and with coconut fiber.

1.3 Structure

This paper is divided into five chapters, organized as follows:

The first chapter is introductory in nature and has the purpose of framing the thesis, referring to the objectives of the work and a brief description of the structure of the dissertation.

The second chapter presents the information from existing studies, concerning natural fibers, fiber-reinforced composites, additive manufacturing and mechanical tests, that is, it describes all the choices made throughout the work.

The third chapter presents a paper that will be published by studying the tensile and flexural strength of flax fiber reinforced composites and the respective references.

The fourth chapter presents a paper that will be published by studying the tensile and flexural strength of composites reinforced with coconut fibers and the respective references.

The fifth chapter presents the main conclusions resulting from the development of this work and some suggestions for future work are made.

Finally, you will find the references used.

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Chapter 2

State of the Art

This chapter presents the study of some important theoretical concepts for the correct understanding of the study developed throughout the work. First, a brief description of fiber-reinforced composites will be presented, followed by fiber types, polymer properties, and the importance of additive manufacturing and finished with mechanical tests.

2.1 Composites

Since the dawn of civilization, man has used natural fibers. With the constant evolution and change imposed on human beings, they have been adapting and giving new applications and uses to these fibers. Currently, the world is very dependent on fossil fuels, and because of this, there has been an increase in pollution and a greater concern with environmental impact, and therefore, it is necessary to replace polluting materials by renewable materials, and ecological, to get around this problem. Thus, there have been several studies to find biodegradable materials and processes. Therefore, natural fiber reinforced composites have emerged [1].

Natural fiber reinforced composites (NFRCs) are cheaper than glass and metal reinforced composites because natural fibers are abundant. Natural fiber reinforced composites have mechanical, thermal characteristics and physical properties lower to glass and metal reinforced composites, and therefore, these have been studied, however, in favor of these composites there are their advantages, i.e., they are biodegradable composites, lightweight, with high stiffness in relation to their weight and promising properties that make these composites a potential candidate for textile engineering and specific parts for the automotive industry. Besides all these points favorable to the use of these ecological composites, it was realized that the production of these composites uses little energy and when they reach the end of their useful life it is still possible to recover a large amount of energy [2].

In the last decade there has been an increase in the number of studies on natural fiber reinforced composites and it is expected that more and more studies on these topics will continue to emerge, and these studies have been mostly in the automotive,

construction, and biomedical fields. However, like any material they have disadvantages. Composites obtained by additive manufacturing, i.e., those that are 3D printed, to have good characteristics, good interface properties of the natural fiber and a good polymer matrix are required [3].

2.2 Types of Fibers

Fibers can be divided according to their origin, natural and synthetic [4]. Comparing natural and synthetic fibers, it can be said that natural fibers have three major advantages over synthetic fibers, they are light in weight, recyclable and biodegradable. In addition to these advantages natural fibers are renewable and have a relatively high strength and stiffness. However, natural fibers absorb moisture, vary in quality, and have low thermal stability [5].

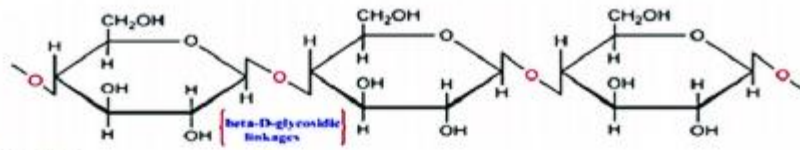
Natural fibers can be divided into three groups, depending on their sources, i.e. animal, vegetable, and mineral. Vegetable fibers are the most accepted by industry and are being increasingly studied with the aim of replacing synthetic fibers. This is due to the fact that in addition to the advantages already mentioned of natural fibers, natural vegetable fibers have a short growing season, are inexpensive, have unlimited availability, and are environmentally acceptable [6]. Table 1 presents the natural fiber division [4]:

Table 1 - The natural fiber division [4].

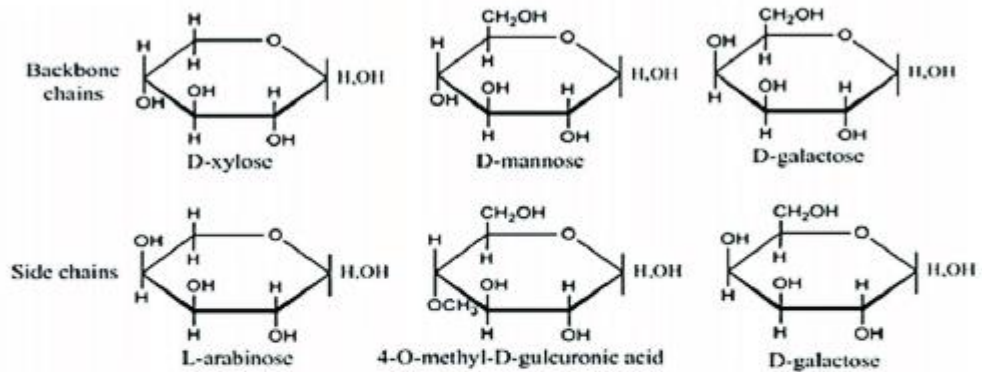
Vegetables	Wood	Soft and hard wood (e.g. eucalyptus)
	Grass/Junco	Corn fiber and bamboo
	Fruit	Coconut
	Stalk	Fiber from rice, wheat, corn and oats
	Fibrous Material	Canamos, kenaf, linen, sisal fiber
	Sheets	Cotton and coconut
Animal	Silk	Hair from goats, sheep and horses
	Coat / Wool	Sheep Wool
Mineral		Glass and graphite

Natural fibers (animal, vegetal and mineral) have in their composition 4 main elements, which are lignin, cellulose, pectin and hemicellulose [7], in which their chains can be visualized in Figure 1 [4].

(a) Cellulose



(b) Hemicellulose



(c) Phenols in Lignin



(d) Pectin

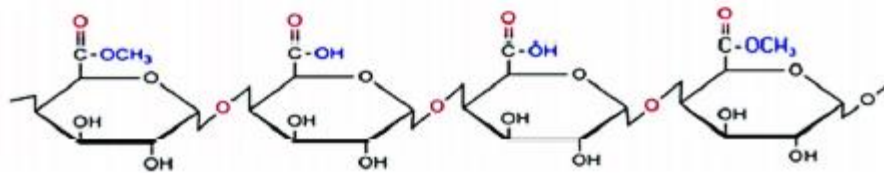


Figure 1 - Composition 4 main elements [4].

Cellulose is the most abundant organic polymer on the planet [8, 9]. This polymer has several -D-glucose (C₆H₁₁O₅) subunits that are linked by glycosidic interactions at the C1 and C4 carbon sites. Cellulose can be found in microcrystalline and amorphous form, with high and low order sites, respectively [10]. Cellulose is a robust material and resistant to oxidizing and reducing agents. However, cellulose is vulnerable to hydrolysis with acid to produce water-soluble, biodegradable sugars when subjected to chemical and solution treatments [11].

Hemicellulose is the second largest biomolecule with abundance in the world, and is composed of polysaccharide subunits, consisting of a branched-chain polymer of carbon-ring sugars. This molecule is the polymerization of carbon ring carbohydrates that are highly branched. Unlike cellulose, it has a higher degree of polymerization and has

shorter chains. It is a hydrophilic molecule and dissolves in alkaline media [12, 13]. This molecule can create bonds to the cellulose and lignin elements of the cell wall, which creates increased fiber stiffness and flexibility [14].

Lignin is a complex nanocrystalline molecule, since it contains functional groups such as hydroxyl, methoxy and carbonyl. The molecular composition of lignin is still unclear, however it is known that lignin is generated within a plant cell wall and is composed of aromatic units, is an insoluble molecule in most solvents [4]. This molecule is a phenolic molecule with a high molecular weight and is often resistant to microbial decomposition and is not broken down into its constituent sub-monomers [10, 15–17]. This molecule increases the plant's rigidity because it is hydrophobic, which makes it act as a compatibilizer between cellulose and hemicelluloses [18, 19].

Pectin is composed of heteropolysaccharides known as glycosaminoglycans, each containing a different amount of methyl ester. The basic pectin, shown in Figure 1 is homogalacturonan (HG) which is an unbranched polymer of galacturonic acid bound [20].

Natural fibers are composed, essentially, of cellulose microfibrils, which are suspended in a matrix of amorphous lignin/hemicellulose composition, thus giving the fibers their chemical composition. The lignocellulose cell wall is a naturally occurring complex structure with a spiral shape containing various biochemical combinations [21]. Lignocellulose is the molecule responsible for the chemical conjugation of plant fibers (cellulose, hemicellulose, and lignin). The amount of these components is different from plant to plant, since it changes due to age, species and also because in the same plant, the different parts are chemically different [22]. Therefore, there are several factors that alter the chemical structure of cellulose fibers, such as the topographic aspect where the plant was grown and the variation of climate in that area [23].

Climate and soil conditions, as well as plant species and age, cause the chemical composition of natural fibers to vary. This causes the fibers to have different chemical, thermal, and mechanical properties. It can be admitted that cellulose is responsible for giving the fiber strength, hemicellulose controls the biological and thermal processes, and lignin is responsible for UV rays destruction and fire resistance [14, 24, 25]. Another characteristic that has a significant impact on fiber characteristics is the fibril angle, that

is, the alignment of the microfibrils in relation to the cell axis. This angle is responsible for fiber stiffness [26].

It is possible to establish a relationship between the microfibrillar angle and the Young's modulus of the fibers, (Figure 2) [27].

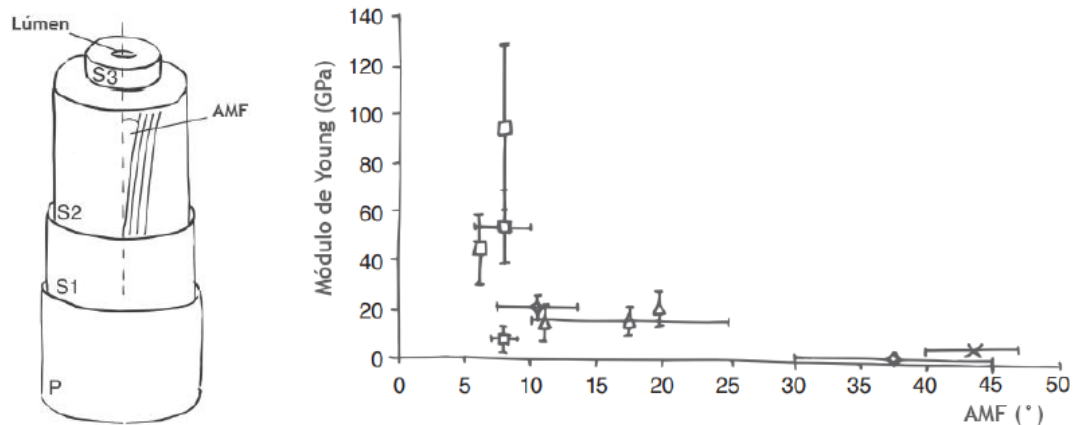


Figure 2 - Microfibrillar angle [27]

It can be seen that for smaller angles, between 6° and 10° of the micro-fibril, high stiffness values result and vice versa. This can be explained by the high orientations of the load-carrying cellulose structures in the direction of load application [27].

There are several techniques to improve the characteristics of natural fibers, however this topic is still being studied and developed. Improving for example the mechanical properties of a fiber in one area of application, may not be equivalent in other areas [4].

2.2.1. Surface modification

Modifying the morphological behavior of the surface of the natural fibers before use to make better bonding at the interface between the matrix and the fiber and also allows you to avoid the shortcomings of natural fibers. It also allows the insufficient layer between the fiber layer and the matrix to be controlled, so that the absorption of water and moisture is controlled [6,28].

2.2.2. Physical Techniques

These techniques have been the most used by man for centuries, for the creation of cuttings, clothes, baskets [6]. In the present study, when coconut fibers were used to

perform the tensile and flexural tests, a net arrangement of the natural fibers was tested to buy with the horizontal arrangement of the fibers in the specimens.

2.2.3. Chemical Techniques

Chemical techniques use chemical reagents to alter the interconnection and surface properties of natural fibers, with the goal of altering the alkalinity and acidity of the fibers by creating a consistent, flexible layer on top of the natural fibers on the fiber surface [29].

2.2.3.1. Alkaline treatment

The treatment of natural fibers with sodium hydroxide (NaOH) is very common, as it allows the chemical composition of the cellulose polymer to be altered by causing a change in the alignment of the cellulose, thus resulting in the formation of an amorphous layer. In this way greater accessibility to substances penetrating the skin is achieved, and the cellulose macromolecules are secreted by large amounts in the amorphous zone with water molecules due to the spaces between them. The hydroxyl groups (OH) located between the molecules are broken down and combine with the water molecules, thus flowing out of the fiber [6]. It also removes a percentage of the hemicellulose, lignin, and pectin, making the surface of the fiber clear and uniform. With this treatment, it is possible to improve the mechanical and thermal characteristics [30, 31].

2.2.4. Coconut fiber

Coconut fiber (Figure 3) is a fiber rich in lignin and is therefore a fiber with worse properties when compared to cellulose-rich fibers such as sisal. Coconut fiber has a high creep resistance of 15-40% and has a thin aliphatic layer. The wax layers consist of long-chain fatty acids and their condensation products. Coconut fibers have a high tensile strength, which leads to better stress compatibility between fiber and matrix. Coconut fibers have a low thermal conductivity and low density. There have been several studies using coconut fiber as a natural fiber for reinforcing composites, in which the results have been promising, and indicate that coconut fiber can be used as reinforcement for composites with low loading stresses. Coconut fiber is harvested from the coconut palm and is an abundant waste product. Large amounts of this agricultural waste are left in the fields and then burned, generating a negative environmental impact. Therefore, by using these fibers for making reinforced composites, it is also possible to reduce the environmental impact generated by the agricultural sector [32–34].



Figure 3 - Coconut Fiber.

2.2.5. Flax Fiber

Flax fibers (Figure 4) are among the best natural fibers currently considered because of their tensile strength, and their abundance. Several tests indicate that PLA composites reinforced with flax fiber have a Young's modulus of approximately 6.52 GPa, a value close to the same composite reinforced with glass fiber with values around 7.76 GPa [35]. The central zone of these fibers consists of pectin and small amounts of lignin. The fiber is composed of a primary cell occupying about 10% of the fiber diameter, consisting essentially of cellulose microfibrils surrounded by a matrix of hemicellulose, pectin, and a small portion of lignin [36, 37]. The remaining 90% is considered a secondary cell wall, which is composed of three microfibrillar layers of cellulose, bounded by pectin and hemicellulose. The structural differences between the primary and secondary layers are quite important in the mechanical and surface properties [38, 39].

Using the alkaline treatment on these fibers allows the removal of amorphous polysaccharides with hemicellulose, pectin, and lignin. It reacts with the cellulosic components creating large cellulose networks, which can convert into new cellulose structures [40].



Figure 4 - Flax Fiber.

2.3. Polymers (PLA)

Natural fiber reinforced composites have become an area of increasing interest because, besides being ecologically feasible, they present desirable properties and performance at a reduced cost. These composites show promise, with future potential, however various materials have been studied in order to achieve long-term stability and performance. Various synthetic polymers reinforced with natural fibers have been studied because they are abundant, recyclable, and relatively inexpensive. Interest is growing in thermoplastics for high-performance engineering applications [41, 42]. These reinforced synthetic polymers are not biodegradable, and therefore still present problems in terms of environmental compatibility. In this context, the solution is to replace synthetic polymers with biodegradable polymeric matrices [39, 43].

Biodegradable composites, then, come to solve the environmental problems caused by synthetic polymers. These composites contain matrix materials derived from agricultural and forestry raw materials and are reinforced by cellulose fibers [44]. The disposal of these composites in life fiber, has no concerns, because unlike synthetic composites, they have unique benefits such as being recyclable, renewable and biodegradable, low density and lower cost. These biocomposites are non-toxic, so they are safer to use than synthetic composites and therefore have a wide range of applications, such as in automotive, aerospace, construction, biomedical, defense, packaging, sporting goods, among others [45, 46].

There are several natural bio polymers such as starch, soy resin, gelatin, polyhydroxybutyrate (PHB), polyhydroxybutyrate (PHBV) and poly lactic acid (PLA), which can be used to form a matrix for a biocomposite. Among all these, PLA has been widely studied, as it is a viable option to replace synthetic polymers that are derived from petroleum. This polymer is biodegradable and renewable and is obtained from farms, from sugar beets, saccharin, corn, etc. PLA has good mechanical strength, good rigidity, and can be manufactured by traditional methods. The life cycle of PLA requires only one-tenth of the fossil energy compared to synthetic polymers. Thus, PLA is an ideal choice to produce biodegradable composites [44].

Natural fiber-reinforced polymers have been increasingly studied as the perfect pairing when it comes to green composites. PLA can be synthesized by condensation of lactic acid and can be produced at a capacity of 140,000 tons per year. Several mechanical

tests have been performed on PLA reinforced natural fibers, such as kenaf, jute, flax, abaca, hemp, etc. [47].

The mechanical properties of natural fiber reinforced composites have a major impact in determining the structural applications for the composites. There have been several studies of PLA composites with various natural fibers in which the fiber parameters have influenced the results. The application of sisal fiber at 20% by weight of a PLA composite improved the tensile and flexural properties up to 80.6 MPa and 249 MPa, respectively [48]. When sisal was increased by 20 to 40%, the tensile strength increased to 352 MPa [11]. Flax fiber at 20% by weight, showed tensile strength up to 76.2 MPa [11] among other studies.

There are various methods to manufacture the PLA-based composites, such as injection molding, injection and extrusion molding, extrusion compression molding, hot pressing, layer stacking and additive manufacturing [44].

The performance of natural fiber-reinforced composites is determined by a combination of factors derived from the properties of the fibers used, the matrix (base composite), and the interfacial interaction between fiber and matrix, for example physical bonding and chemical bonding. Despite the great advantages of natural fibers already listed, they also have characteristics, which are considered an obstacle to the production of high-performance composites and their applications. These obstacles are mainly linked to the heterogeneous characteristics of the fibers (variation in cell wall structure, composition, and geometry), leading to a change in the mechanical properties [49, 50].

2.4. Additive manufacturing

Additive manufacturing of composites is a technique that is increasingly being used, this technique can also be referred to as 3D printing. In this method a filament comprising the matrix and the reinforcement is used. The filament is guided through a heated nozzle, which with the help of a feed roller melts the material and then deposits it on a worktable, layer upon layer. After the material solidifies, the last deposited layer fuses with the previous layer, and so on (Figure 5) [51].

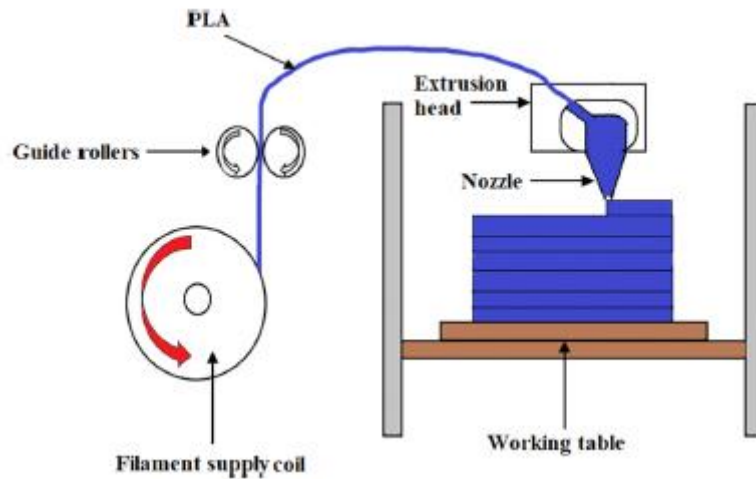


Figure 5 - Additive manufacturing [51].

Currently there are still few studies regarding 3D printing of natural fiber composites with PLA being the matrix material. Additive manufacturing to create composites is still in its infancy, so there has been a lot of research conducted to understand the process.

From additive manufacturing of fiber-reinforced PLA composites, one can derive several advantages, such as easy and fast manufacturing, low thermal degradation, cheaper tooling, better control of fiber volume and direction. However, it also has some disadvantages, which are mainly related to the material properties being inferior compared to conventional methods, hence further studies are needed to optimize the process. Nozzle clogging also occurs regularly [52].

Fused deposition modeling is one of the simple 3D printing techniques that is widely used because there are several polymers available, such as PLA. It is also possible to directly manufacture the composite and the natural fiber together with the filament composite, as shown in Figure 6 [53].

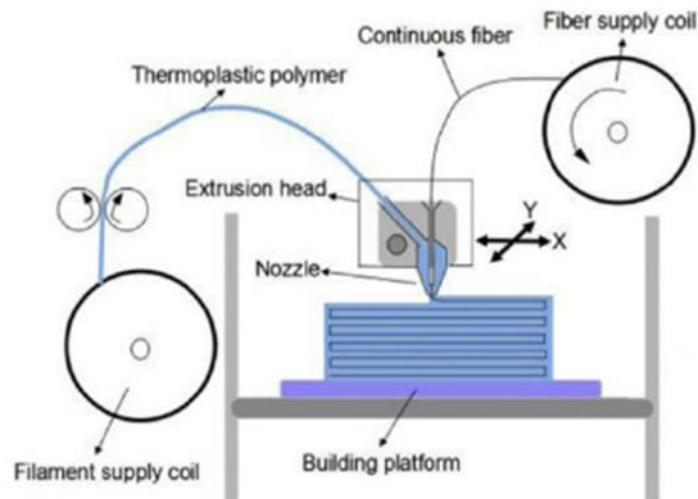


Figure 6 - Additive manufacturing of natural fiber with composite [53].

With this process it is possible to obtain a natural fiber reinforced composite with a percentage between 40 and 55% natural fiber content. In this system there are two rolls, one of the rolls has the filament and the other has the natural fiber. The way of working is the same as the previous system, however the fiber is inserted directly with the composite, leaving the aspect that can be seen in Figure 7 [53].

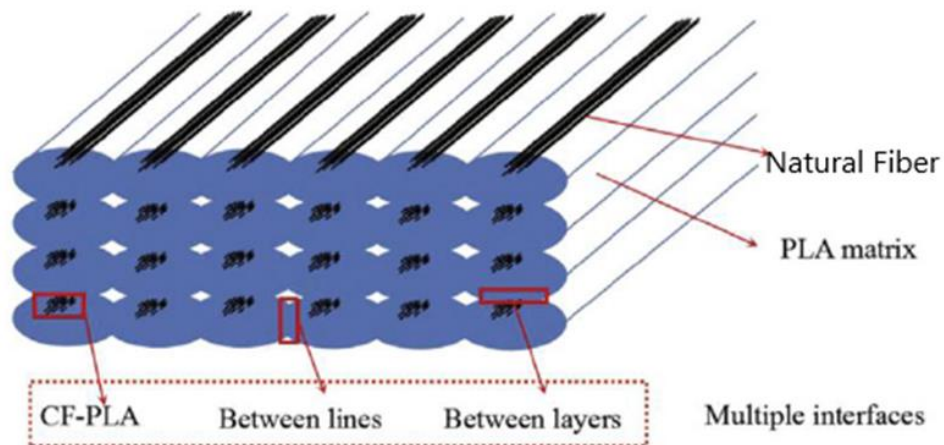


Figure 7 - Layers of the reinforced composite [53].

The properties of natural fiber-reinforced composites obtained by additive manufacturing tend to be lesser to the properties obtained in the injection molding process, since the additive manufacturing process does not use compressive stress during composite production and thus the contact between the fibers and the base matrix (polymer) may not be fully effected, resulting in a poor interface and consequently a poor bonding face. However, the thermal degradation of the natural fiber reinforced

composites produced by additive manufacturing was similar to the composites obtained by injection molding. Nevertheless, it was found that the shrinkage of the composites is lower when they are produced by additive manufacturing [54, 55].

2.5. Mechanical tensile and bending tests

To perform the mechanical tests two standards were taken into account, ATSM D638.14 for tensile tests and ISO 14125 for flexural tests [56, 57]. There are several parameters that create influences on the test results of composites, such as test speed, measurement length, cross-sectional area, number of specimens, and component type.

2.5.1. Tensile test

In tensile tests the objective is to determine the tensile strength of composites. This mechanical property is one of the most important for the design of various materials.

To obtain the material results, it is necessary to create the specimens with their own geometry and apply the loading data to the universal testing machine. The geometry of the specimens must be made in such a way that when they reach failure, this occurs in the desired location, i.e., in the shortest section. The most accepted geometry for these specimens is the "dog bone" or "bell" geometry, i.e., specimens with a conical width like that of ASTM D 638.

The fibers must be correctly aligned, as this is very important to obtain the correct elastic properties. An incorrect alignment of the fibers in relation to the test axis can result in an error of about 30%. Before placing the specimen in the machine, it is necessary to accurately measure the cross-sectional area and its length. After this you can place the specimen in the universal testing machine, placing the ends in the clamps of the machine.

Unidirectional composites exhibit linear behavior. However, it should be noted that at the beginning of the test some amount of nonlinearities may be present, which can be attributed to slip phenomena, machine backlash, etc.

2.5.2. Flexural test

When you are looking at bending tests there are several tensile, compressive and shear stresses. So, the flexural strength cannot be used directly in the design of composites. The main purpose of the bend test is to determine the maximum bending force. Two methods can be used to perform the bending tests, three-point bending test and four-point bending test.

The three-point bending test uses a flat beam-shaped specimen of constant rectangular cross section. The dimensions of the specimen are not exact, only an appropriate ratio of span thickness (L/h) should be chosen. This ratio must be large enough so that the failure occurs in the bending of the specimen [58].

The calculation methodology for each of these tests is presented in chapters 3 and 4, as well as the characteristics of the specimens used, through the ISO 14125 standard.

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Chapter 3

Mechanical properties of PLA specimens obtained by additive manufacturing process reinforced with flax fibers¹

Abstract

The polylactic acid (PLA) is one of the most used materials in additive manufacturing processes, however its mechanical properties are very limited for most practical applications. So, to overlap these limitations, many researchers have been reinforcing it with fibers. In present work, the authors developed PLA composites reinforced with flax fibers. To analyse the influence of fibers in the composite, was created a Taguchi L18 array where the control factors were the extruder temperature, fiber percentage, fill percentage of the specimens, and whether the flax fiber had chemical treatment. As for the treatment of flax, when applicable, it was treated with sodium hydroxide (NaOH). The extruder temperatures used were 190°C, 200°C and 220°C. The percentage of fiber varies in 10, 15 and 20 fiber strands. The filling percentage of the specimen varies between 25%, 50% and 100%. This was followed by a study of tensile and flexural strength for the reinforced composites. The optimal combinations for increased tensile and flexural strength were obtained with the following parameters for tensile: with/without chemical treatment, with a temperature of 200°C, 10 fibers and a filler percentage of 100%. For bending the parameters was: with chemical treatment, a temperature of 190°C, 10 fibers and a fill percentage of 100%. With the confirmation tests a value for the maximum tensile stress of 48.43 MPa and for bending of 71.90 MPa was obtained.

Keywords: composite with natural fibers; flax fibers; PLA; additive manufacturing; mechanical properties.

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3.1 Introduction

In last decade, additive manufacturing processes have been increasingly used in different applications [1]. The applications of these manufacturing techniques can range from prototype manufacturing (first applications) [2] to industry [3,4], through leisure [5], scientific research [6,7], among other applications [8,9]. There are many additive manufacturing processes [10], however, one of the most common is the fused deposition modelling (FDM). FDM, also known as fused filament fabrication, is a process within the field of material extrusion. FDM employs filaments made of thermoplastic polymers and creates parts layer by layer by selectively depositing melted material along a predetermined path [10]. The most popular thermoplastic polymers used in FDM are the PLA (polylactic acid) and ABS (acrylonitrile butadiene styrene). ABS is a polymeric material that derive from petroleum in their composition have volatile organic compounds that can cause damage to the environment and human health [11]. On the other hand, PLA is a biodegradable polymer, with much technological interest due to its applications in the environmental field [12]. It is a type of impact-modified filament for the 3D printer, which is sustainable, does not use volatile organic compounds, and allows the final product to have an accelerated degradation time by the action of humidity, temperature, light, and soil microorganisms, thus decreasing its mechanical properties and its useful life. As this is a thermoplastic polymer, which comes from renewable sugar-based raw material, it may be an alternative to the use of non-biodegradable polymers or polymers with long-term degradation [13,14]. Despite the advantages of these materials like good adaptability to the FDM process, low cost and the obtained parts have good resolution [15], the mechanical strength is relatively low [16] for more demanding applications, so it would be interesting to improve some mechanical properties, like, tensile and flexural strength and one possibility is to reinforce the parts with fibers (natural or synthetic).

Many automotive components use synthetic fibers for strength, however, with the increasing demand for sustainability and due to the energy crisis, it is necessary to conduct a study in order to circumvent the causes of these products and find a sustainable way to replace them. Natural fibers are fibers that are not synthetic or manufactured. These fibers come only from animals or plants [17]. Animal fibers consist of proteins (wool or silk), while plant fibers consist of cellulose [18]. Currently, natural fibers that originate from plants are the most widely used because they are suitable for use in composites with structural requirements. Still, plant fiber can be grown in many countries

and can be harvested after short periods of time. In addition to cellulose, natural fibers are composed of lignin, hemicellulose, pectin, and waxes, and can be considered as natural composites containing mainly cellulose fibrils embedded in a lignin matrix. The nature of the cellulose and its crystallinity play an important role in the reinforcing efficiency of the natural fiber. The cellulose fibrils are aligned along the length of the fiber, ensuring maximum tensile and flexural strength and providing stiffness [19].

Natural fibers have unique characteristics such as abundance, non-toxicity, high performance, versatility, and easy processing at low cost. The natural fiber reinforced polymer composites (NFPCs), have several applications, besides the automotive industry, also in the construction industry due to their strength, low density, biodegradability and high lifetime. The fibers most used in industrial applications are flax, knaf and hemp because of the fiber's strength properties [20]. The properties of natural fibers vary, as they depend on the type of fiber, its source, and the moisture conditions. These properties depend on the fiber composition, the microfibril angle (angle of orientation of the microfibril in relation to the main fiber axis [21]), structure, defects, cell dimensions, physical properties, chemical properties, and also the fiber matrix [17]. Flax is one of the oldest cultivated plant species in the world, a member of the genus *Linum* in the family Linaceae, *Linum usitatissimum* L being the most common species among the 298 different species that exist. Flax fibers are found near the stem and are the mechanical support of the plant that is very thin [22–25].

Flax fiber is considered one of the strongest fibers, because it has a very complex structure. These fibers are made up of a series of polyhedra that form overlapping elementary fibers over a considerable length, held together by an interface consisting mainly of hemicellulose and pectin. The typical diameter of an elemental flax fiber is between 10 and 15 μm , although technical flax fibers range between 35 and 150 μm [12].

However, since natural fibers have some limitations, the formation of the composite and the treatment parameters must be controlled in order for the product to improve its properties [13]. Due to these limitations it is necessary to proceed to the use of treatment processes that can be chemical or physical that while increasing the price of the fiber also improves the adhesion properties of the interface between the fibers and the matrix and decreases the absorption of water by the fibers [14,26,27]. These processes can be considered as modifiers of the properties of these fibers [28]. One of the most used chemical treatments is the alkaline one, in which the cracks are submerged in an alkaline

solution, namely NaOH (sodium hydroxide), for a relative period. This increases the fiber's surface roughness and improves its mechanical properties. At the level of chemical bonding with the matrix material, it is possible to expose more cellulose on the fiber surface [29,30]. The most important modification that is made by this treatment is the breaking of the hydro-gen bonds in the lattice structure, thus causing the surface roughness to increase. It also removes a certain amount of lignin, wax and oils that cover the outer surface of the fiber cell wall, exposing the short length crystals and depolymerizing the cellulose [31].

For the substitution of products that do not come from renewable sources, it is possible to use biodegradable polymers, i.e. products from renewable sources, such as compo-sites with natural fibers [32]. Natural fibers can be added to PLA to reduce environmental and production costs, thus allowing to increase the biodegradation process, minimize pollution and, at the level of mechanical properties, they will be more resistant. [33]. With the advancement of 3D printing, PLA has become a widely used material due to its ease of printing, low cost, and good moldability. 3D printing is very interesting in the area of biomaterial development [34].

The study presented in this paper, aims to compare the tensile and flexural strength of flax fiber reinforced composites by analyzing the influence that the amount of flax fiber, the temperature, the percentage of filler and the presence or absence of NaOH treatment have on the mechanical strength of the composites.

3.2 Material and Methods

In this chapter you can see all the materials and methods used in the course of the laboratory tests, regarding the making of the specimens, the chemical process, and the tensile and flexural tests.

3.2.1. Design of experiments

Based on literature referred in the previous section, there are different manufacturing parameters that have a great influence in the properties of obtained products. So, quality of the parts made by additive manufacturing have a strong influence by nozzle temperature and the filling level. Normally these specimens inside are hollow, and it was decided to vary this cavity by changing its filling percentage by 25%, 50% and 100%. In contrast, the mechanical properties of NFPCs suffer an important variability for different

fiber volume fraction (or fibers percentage) and with the fiber surface treatment. For these reasons, the authors of this work decided to use these parameters as control factors with different levels. In Table 2 is presented the control factors with the respective levels.

Table 2 - Control factors of the composite.

Symbol	Control factor	Level 1	Level 2	Level 3
A	Fiber surface treatment	NaOH treatment	No treatment	
B	Nozzle temperature	190°C	200°C	220°C
C	Number of strands	10	15	20
D	Filling level	25%	50%	100%

With the control factors and their respective levels previously defined, it was possible to create a Taguchi L18 orthogonal array (Table 3). This array was used for the implementation of both groups of experimental tests.

Table 3 - Taguchi L18 orthogonal array.

Test number	A	B	C	D
	Fiber surface treatment	Temperature	Fibers Percentage	Filling level
1	1	1	1	1
2	1	1	2	2
3	1	1	3	3
4	1	2	1	1
5	1	2	2	2
6	1	2	3	3
7	1	3	1	2
8	1	3	2	3
9	1	3	3	1
10	2	1	1	3
11	2	1	2	1
12	2	1	3	2
13	2	2	1	2
14	2	2	2	3
15	2	2	3	1
16	2	3	1	3
17	2	3	2	1
18	2	3	3	2

3.2.2. Specimens manufacturing

To determine the tensile and flexural strength it must be manufacture specimens to tensile tests and to flexural tests and was used the standards ASTM D638.14 [35] and ISO 14125 [36], respectively. So, the geometries and dimensions of these specimens must

agree with these standards. In Figures 8 are represented the drawing of the two types of specimens. Table 3 shows the values of specimens dimensions.

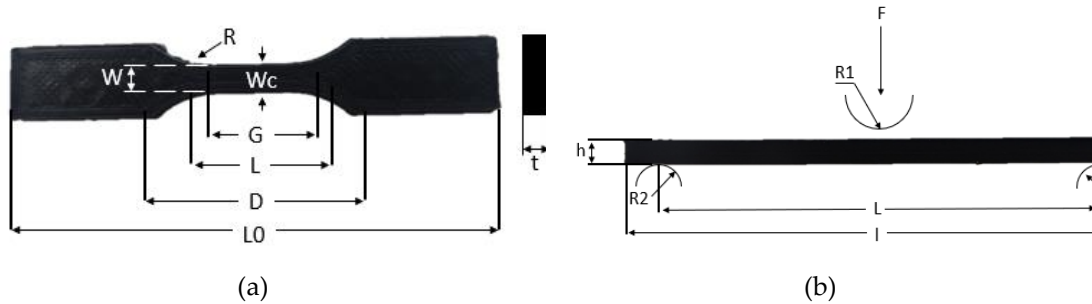


Figure 8 - (a) Tensile specimen geometry in agreement with ASTM D638.14 standard and (b) Flexural specimen geometry in agreement with ISO 14125 standard.

Table 4 - Tensile (a) and flexural (b) test piece dimensions. Dimensions in mm.

Tensile test piece dimensions [mm]		Flexural test piece dimensions [mm]	
D	30	F	-
G	10	h	4
L	19	I	80
L0	63	L	64
R	13	R1	10
t	4	R2	10
W	4		
W0	10		
WC	4		

(a)

(b)

Briefly, the main materials/equipment used to manufacture the specimens are:

- EasyFil PLA;
- Anycubic 3D printer;
- Linen fibers;
- MDF mold;
- Scissors;
- 3M tape.

The flax fibers are used to reinforce the 3D printed specimens. On the other hand, to manufacture the specimens, was chosen a biodegradable thermoplastic, PLA (EasyFil PLA), from FormFutura, which has the characteristics of being easy to print, low deformation, good adhesion between. To spatially position the fibers in the defined locations, it was necessary to develop molds. These molds were manufactured from

wooden (MDF) and boards were cut using a cutting laser machine (GCC X252). Figure 2 shows the molds used in this work (mold (a) use in tensile specimens and mold (b) used in flexural specimens). The fibers were fixed in the grooves of the molds and the mold-fiber system were placed on the table of the 3D printer (Anycubic 3D printer) where the PLA deposition takes place.

The process it began by joining each of fiber with small knots, and wrapping them around the mold according to the number of fiber strands that would be wanted, i.e., with 10, 15, or 20 strands. After completing the turns, the tape and scissors were used to fix them so that they would not move, since in the mold created are inserted grooves to put the fibers in the right position. When this is finished, the fused PLA is placed, and the program previously inserted into the 3D printer is started to manufacture the specimens. Printing is started, and when it reaches 50% of the specimen, the printer is paused and the mold fibers are placed. To keep the fibers stretched and fixed, glue tape was used, thus al-so preventing the printer from damaging the fibers. Printing continues for the remaining 50% of the specimens. After all this process, which takes about 2 hours for tensile tests and 4 hours for bending tests, it is necessary to let the specimens and the machine cool down to remove them (Figure 10 (a) and (b)), finishing the process by wrapping the 6 and 12 specimens, tensile and bending tests respectively, in tape and identifying them with their characteristics.

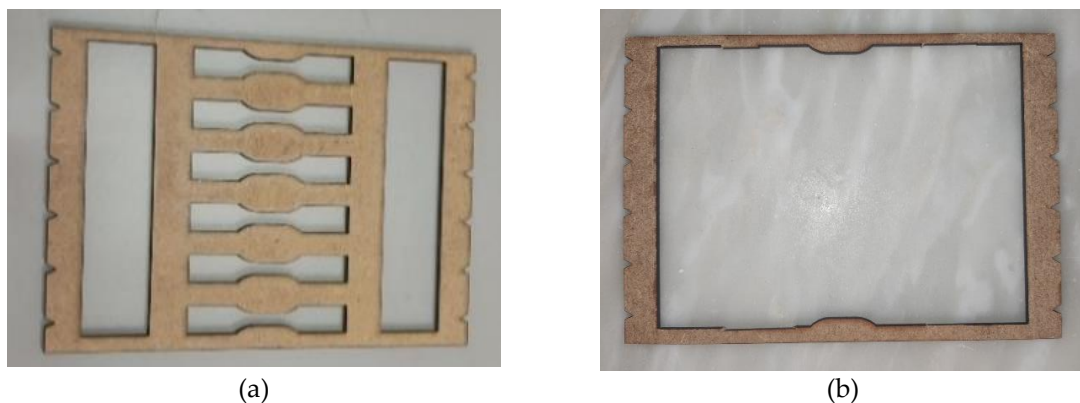


Figure 9 - Molds used for tensile specimens (a) and flexural specimens (b).

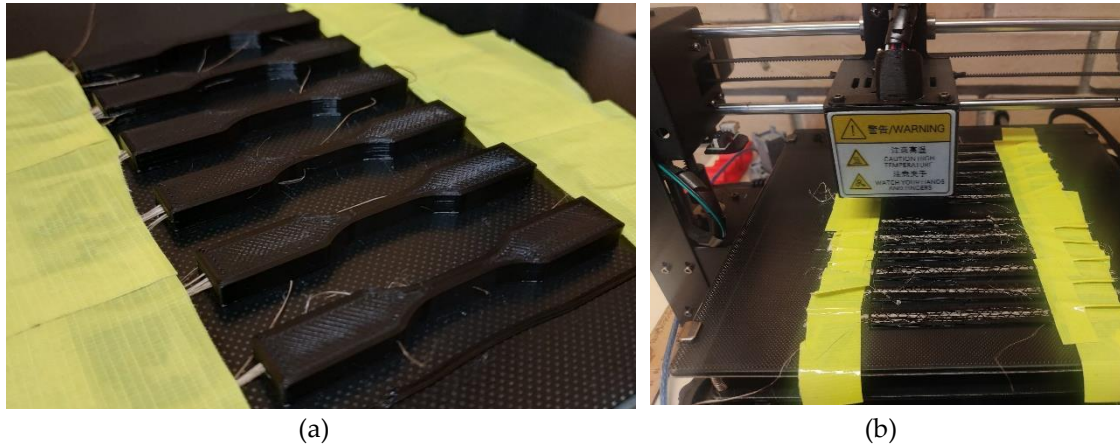


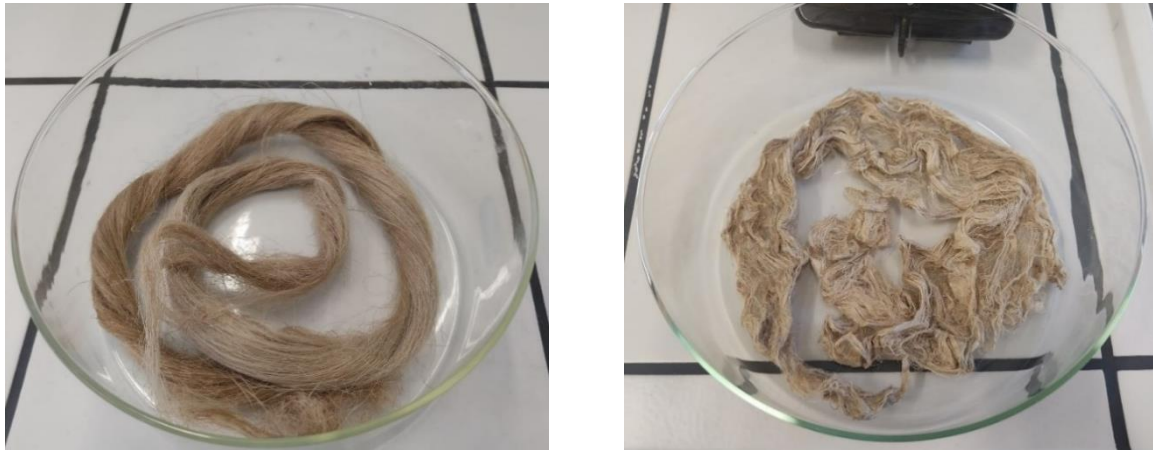
Figure 10 - Printed specimens (a) and labeled specimens (b).

3.2.3. Chemical surface treatment

Regarding the chemical surface treatment, the following materials had to be used:

- Flax fibers (Figure 11 (a));
- 2 L of 5% aqueous NaOH solution;
- 2 L of 5% aqueous acetic acid solution;
- 2 L distilled water;
- Scientific Series 9000 Oven;
- 2 containers.

To improve the surface properties of the flax fibers, they were subjected to a sodium hydroxide treatment. The flax fibers were immersed in a 5% sodium hydroxide (NaOH) solution for 3 hours at room temperature, washed with 5% acetic acid to neutralize the NaOH, washed with distilled water to remove all residues of acetic acid, then dried in a 120°C oven for 2 hours, and dried in a natural environment for 24 hours, the result can be seen in Figure 11 (b).



(a)
Figure 11 - Untreated flax (a) and Treated flax (b).

3.2.4. Tensile tests

For the tensile tests it was necessary to use the following material/equipment:

- Tensile specimens made in previous step;
- Shimadzu universal testing machine;
- Computer.

The tensile test took place in the laboratory of structures and strength of materials. First, the machine and computer were turned on and the data regarding the specimen measurements, the test speed, and what kind of results were desired were entered. To perform the experimental test was used ASTM D638.14 [35] standard, in which the test speed was 10mm/min. Thus, a specimen (figure 12 (a)) was inserted in the machine, placed at 30mm between the clamps (figure 12 (b)).

The tensile strength can be calculated from the maximum load and the transverse area. For the representation of the stress-strain curve it is necessary to determine, for each point, the respective values. The stress is calculated by dividing the applied load by the average cross-sectional area.



(a)



(b)

Figure 12 - Tensile specimen (a) and tensile tests (b).

3.2.5. Bending tests

For the bending test (figure 13) it was necessary to use the following material:

- Bending specimens;
- Shimadzu universal testing machine;
- Computer.

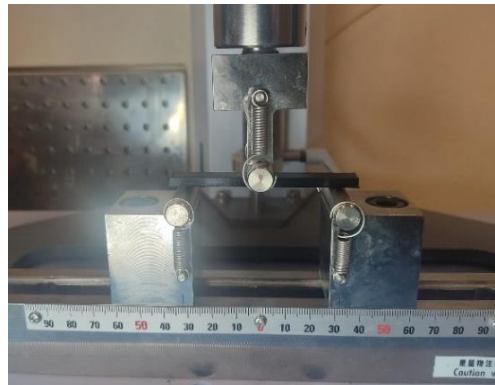


Figure 13 - Bending test.

The bending test took place in the same laboratory where the tensile tests were made, proceeding in the same way, i.e., first the machine and the computer were turned on and the data regarding the specimen measurements, the test speed, and what kind of results would be desired were entered. The standard used was ISO 14125 [36], and a test speed was 1.7mm/min.

The test speed was calculated using the expression:

$$V = \frac{\epsilon L^2}{6h} \quad (1)$$

Where:

V: Test speed (mm/min)

ϵ' : Deformation rate of 1%/min

L: Outer span (mm)

h: Thickness (mm)

Thus, the specimen was inserted in the machine and with the help of the computer the test was started. After finishing all the tests, it was possible to check the graphs and the obtained data.

For the three-point bending test (figure 13) it is necessary to use a test specimen, that is, a specimen in the shape of a flat beam of constant rectangular cross section. In three-point bending, the maximum bending stress occurs at the outer surface of the specimen and is given by:

$$\sigma = \frac{3PL}{2bh^2} \quad (2)$$

Where:

σ : Bending stress at the outer surface (MPa)

P : Load applied at the point (N)

L: Specimen length (mm)

b: Specimen width (mm)

h: Specimen thickness (mm))

The strain is calculated from the equation:

$$\epsilon = \frac{6\delta h}{L^2} \quad (3)$$

Where:

ϵ : Deformation

δ : Deflection - Distance of the lower or upper surface of the specimen in the middle of the span that has deviated from the initial position (mm)

h : Specimen thickness (mm)

L: Specimen length (mm)

3.3 Results

This chapter discusses the experimental results obtained by the tensile and flexural tests.

After analyzing the results obtained using the universal testing machine, stress and strain were calculated, and a graph was drawn for each of the 6 specimens of the 18 tests, obtaining a total of 216 specimens for flax fibers. An analysis of each graph was performed, obtaining the following graphs of the averages for each test.

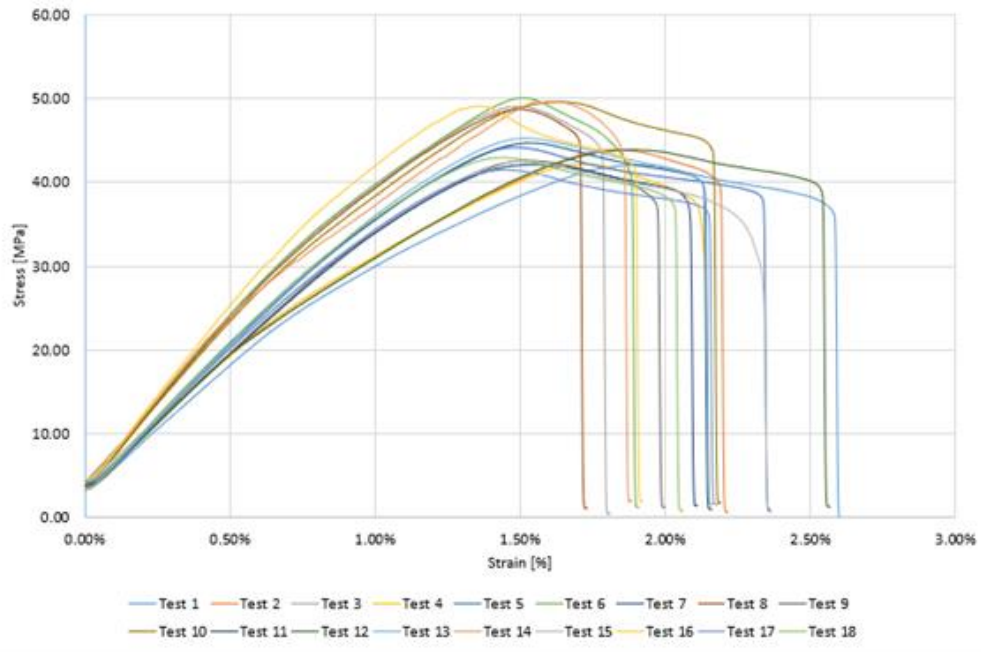


Figure 14 - Stress-strain for tensile strength.

Figure 14 represents the stress-strain curve for tensile strength, showing that test 6 achieve to the best results with a maximum stress of, followed by tests 3 and 8. Test 6 rep-represents the chemically treated specimens, at a temperature of 200°C, with 20 fibers and a fill percentage of 100%. Test 10 represents the specimens without chemical treatment, at a temperature of 190°C, with 10 fibers and a fill percentage of 100%. Test 14 represents the specimens without chemical treatment, at a temperature of 200°C, with 15 fibers and a fill percentage of 100%. In other words, both tests have in common that the filling percentage is total.

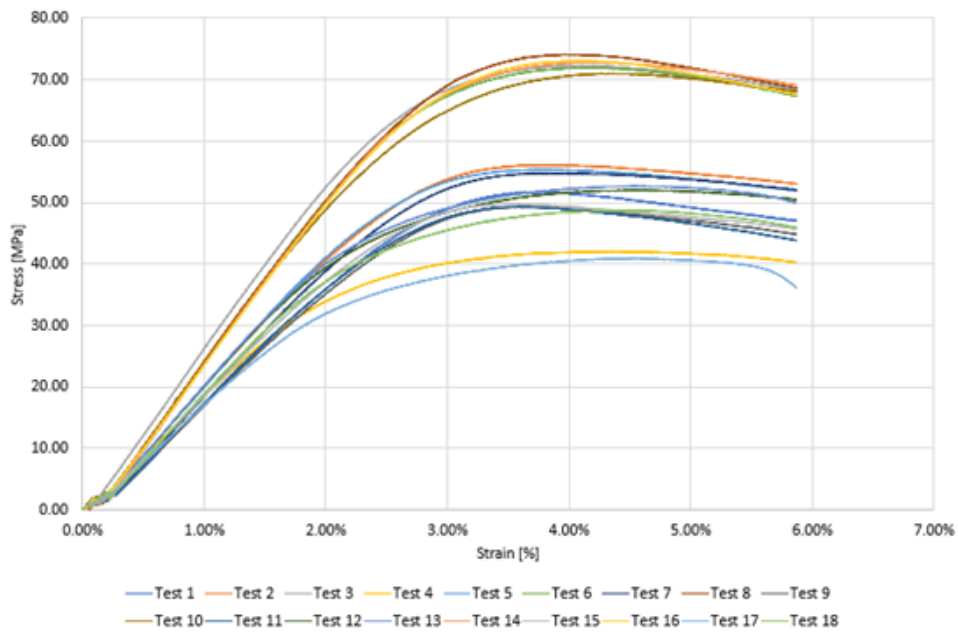


Figure 15 - Stress-strain for flexural strength.

Figure 15 represents the stress-strain curve for flexural strength, showing that test 8 showed the best results, followed by tests 16 and 14. Test 8 represents the chemically treated specimens, at a temperature of 220°C, with 15 fibers and a fill percentage of 100%. Test 16 represents the specimens without chemical treatment, at a temperature of 220°C, with 10 fibers and a fill percentage of 100%. Test 14 represents the specimens without chemical treatment, at a temperature of 200°C, with 15 fibers and a fill percentage of 100%. In other words, both tests have in common only the percentage of filling being in full.

Table 5 shows the mean values of the maximum stresses for the 6 specimens in each test, as well as their standard deviation. As represented graphically, it can be seen that essay 6 obtained the highest maximum tensile stress value for the tensile test with a value of 49.96MPa and a standard deviation of 0.89MPa. For the bending tests, the one with the highest stress was test 8 with a value of 72.94MPa and a standard deviation of 2.09MPa.

Table 5 - Values of the mean maximum stresses and their standard deviation.

EXPERIMENT NUMBER	TENSIL STRENGTH AVAREAGE [MPa]	STANDARD DEVIATION [MPa]	FLEXURAL STRENGTH AVAREAGE [MPa]	STANDARD DEVIATION [MPa]
1	41.99	0.72	51.04	0.78
2	44.02	0.52	54.13	3.38
3	48.25	1.59	72.59	0.79
4	41.99	0.49	44.32	3.96
5	44.01	0.70	54.22	2.79
6	49.96	0.89	71.86	1.13
7	44.55	1.02	52.80	3.52
8	49.16	0.88	72.94	2.09
9	41.86	1.09	46.71	3.68
10	49.55	0.68	70.97	0.50
11	42.11	0.50	46.30	3.81
12	44.41	0.59	53.57	3.05
13	45.03	0.83	54.08	2.85
14	49.25	1.02	72.13	1.71
15	42.69	0.54	48.89	2.75
16	48.85	0.65	72.41	1.69
17	41.09	0.48	41.78	2.75
18	43.26	1.37	50.97	3.82

3.4 Discussion

The experimental results can be converted into a signal-to-noise ratio (S/N). Taguchi suggests using the S/N ratio to determine the quality characteristics that deviate from the desired values.

The Signal-to-Noise ratio developed by Taguchi is a performance measure for choosing the control levels that best handle noise. In this method the term "signal" symbolizes the desired value for the output characteristic, and the term "noise" symbolizes the undesired value. The Signal-to-Noise ratio takes into account the mean and variance, that is, it is the ratio between the mean (signal) and the standard deviation (noise). The S/N equation depends on the criteria for the quality characteristic to be optimized. There are three categories of the quality characteristics in the S/N ratio analysis, which are the lowest better, the highest better and the nominal best. By calculating the S/N analysis you can determine the factor level for the S/N ratio. Regardless of the quality category of the features, a higher S/N ratio will correspond to better quality features. Thus, the level with the highest S/N ratio is the optimal level of the control factors. With the analysis of variance it is possible to see which control factors

are statistically feasible, using the results obtained in the S/N and ANOVA analyses, the optimal combination of control factors and the prediction of their levels. The goal of this study is to maximize tensile and flexural strength, so the quality feature category for the S/N ratio is highest-best:

$$S/N = -10 \log \left(\frac{1}{n} \sum_{i=1}^n \frac{1}{y_i^2} \right) \quad (4)$$

where n is the number of observations and y_i are the observed data [19,37].

The S/N ratios for tensile and flexural strength are shown in table 6.

Table 6 - S/N ratios for tensile and flexural strength.

TEST NUMBER	A	B	C	D	S/N_{ts} ratio [dB]	S/N_{fs} ratio [dB]
1	1	1	1	1	32.46	34.15
2	1	1	2	2	32.87	34.61
3	1	1	3	3	33.66	37.22
4	1	2	1	1	32.46	32.84
5	1	2	2	2	32.87	34.65
6	1	2	3	3	33.97	37.13
7	1	3	1	2	32.97	34.39
8	1	3	2	3	33.83	37.25
9	1	3	3	1	32.43	33.30
10	2	1	1	3	33.90	37.02
11	2	1	2	1	32.49	33.22
12	2	1	3	2	32.95	34.54
13	2	2	1	2	33.07	34.63
14	2	2	2	3	33.84	37.15
15	2	2	3	1	32.61	33.74
16	2	3	1	3	33.78	37.19
17	2	3	2	1	32.27	32.37
18	2	3	3	2	32.71	34.07

In table 6, S/N_{ts} is the S/N ratio for tensile strength and S/N_{fs} is the S/N ratio for flexural strength, where the S/N results for the 18, L18 combinations are represented.

For a higher S/N ratio, the best category is applied in order to maximize the response (tensile and flexural strength). The average S/N ratio for the control factors of levels 1, 2 and 3 can be calculated by averaging the S/N ratios of the corresponding tests. In table 7 and 8, the average S/N ratio for each level of control factor, i.e. the response, is shown.

Table 7 - Mean response table of S/N ratio for tensile strength and significant interaction.

Symbol	Control factor	Mean S/N ratio [dB]		
		Level 1	Level 2	Level 3
A	Fiber surface treatment	33.06	33.07	
B	Temperature	33.05	33.14	33.00
C	Fibers Percentage	33.11	33.03	33.05
D	Filling level	32.45	32.91	33.83

Table 8 - Mean response table of the S/N ratio for flexural strength and significant interaction.

Symbol	Control factor	Mean S/N ratio [dB]		
		Level 1	Level 2	Level 3
A	Fiber surface treatment	35.06	34.88	
B	Temperature	35.13	35.02	34.76
C	Fibers Percentage	35.04	34.88	35.00
D	Filling level	33.27	34.48	37.16

Figures 16 and 17 show the response graph of the S/N ratio for tensile and flexural strength, respectively.

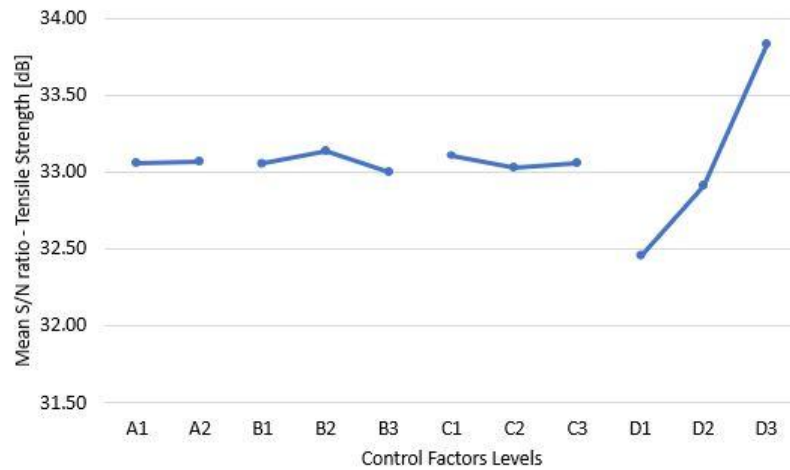


Figure 16 - S/N ratio response graph for tensile strength.

Through the analysis of Figure 16 it is possible to determine the optimal combination to increase the tensile strength of the flax fiber reinforced composite. Regarding the tensile tests the ideal combination is A1/A2, B2, C1 and D3, i.e., with/without chemical treatment, with a temperature of 200°C, 10 fibers and a filler percentage of 100%.

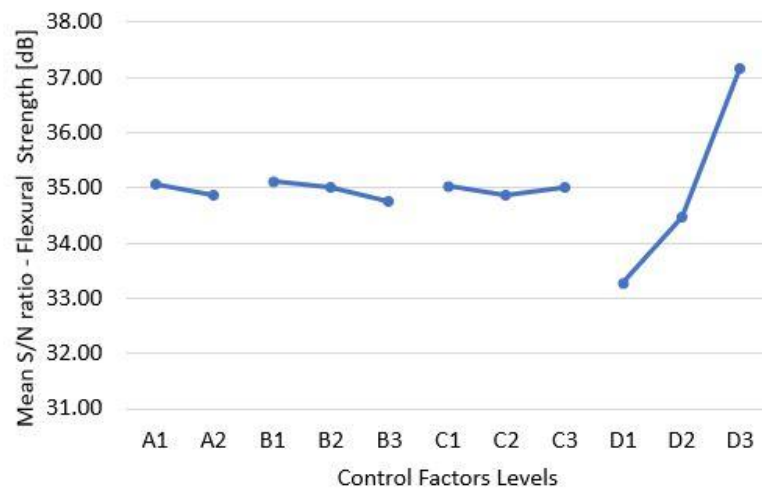


Figure 17 - S/N ratio response graph for flexural strength.

Through the analysis of figure 17 it is possible to determine the optimal combination to increase the flexural strength of the flax fiber reinforced composite. Regarding the bending tests the ideal combination is A1, B1, C1 and D3, i.e., with chemical treatment, a temperature of 190°C, 10 fibers and a filler percentage of 100%.

The relative importance of the control factors for tensile and flexural strength needs to be identified more precisely using ANOVA analysis to determine the optimal

combinations of control factor levels. In Tables 9 and 10, it can be seen that which control factor influences more the tensile and flexural strength values.

Using Tables 9 and 10, you can check the variance results for each control factor, where DF is the degree of Freedom, Adj SS is the sum of squares and Adj MS are the mean squares. The F-test is a statistical tool to check which parameters significantly affect the quality of the characteristics, that is, it is defined as the ratio of the mean square deviations to the mean square error.

Table 9 - ANOVA for tensile strength.

Source	DF	Adj SS	Adj MS	F-Value	P-Value	Contribution [%]
A	1	0.00	0.00	0.00	0.96	0.00%
B	2	0.07	0.03	2.85	0.11	1.12%
C	2	0.01	0.01	0.57	0.58	0.22%
D	2	5.80	2.90	245.99	0.00	96.69%
Error	10	0.12	0.01			1.97%
Total	17	6.00				100.00%

Table 10 - ANOVA for flexural strength.

Source	DF	Adj SS	Adj MS	F-Value	P-Value	Contribution [%]
A	1	0.14	0.14	0.88	0.37	0.29%
B	2	0.43	0.21	1.31	0.31	0.85%
C	2	0.08	0.04	0.26	0.78	0.17%
D	2	47.52	23.76	146.00	0.00	95.42%
Error	10	1.63	0.16			3.27%
Total	17	49.80				100.00%

After analyzing the results of the F-test value, it is possible to verify that for tensile and flexural strength, the most significant control factors are temperature and the filling percentage of the specimen, with the remaining factors varying. That is, for tensile

strength, the percentage of filler was very significant with 96.69%, followed by the nozzle temperature, fiber percentage, and the use or not of fiber treatment, with 1.12%, 0.22%, and 0%, respectively. For flexural strength, the percentage of filler was also the most significant with 95.42%, followed by the nozzle temperature, the use or not of fiber treatment, and the percentage of fibers with 0.85%, 0.29% and 0.17%, respectively.

Confirmation Tests

Given the results obtained, the tests were performed again for the optimal combinations, i.e., for the tensile tests, 3 specimens were made without chemical treatment, with a nozzle temperature of 200°C, 10 fibers, and a filling percentage of 100%. For the bending tests, 3 specimens were also made, but with chemical treatment, a temperature of 190°C, 10 fibers, and a fill percentage of 100%. Only 3 specimens were made because the standard deviation of the previous tests was quite small. Figure 18 graphically represents the optimal combinations of tensile strength.

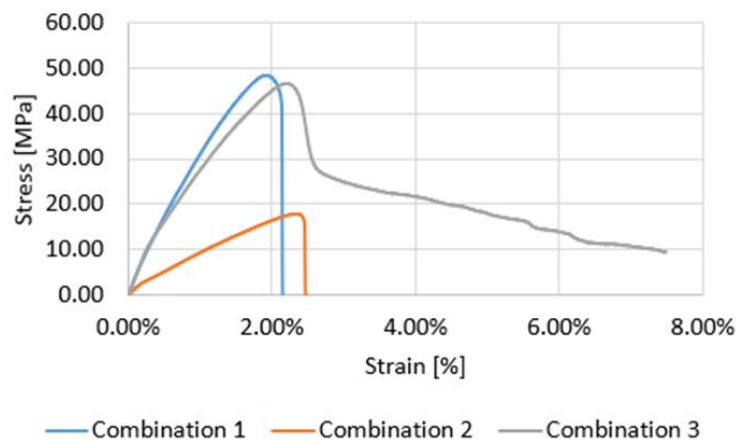


Figure 18 - Optimum combinations of tensile strength.

A maximum stress of 48.43MPa can be observed, i.e., it is very close to the stresses of the best tests performed previously. Figure 19 graphically represents the optimal combinations of flexural strength.

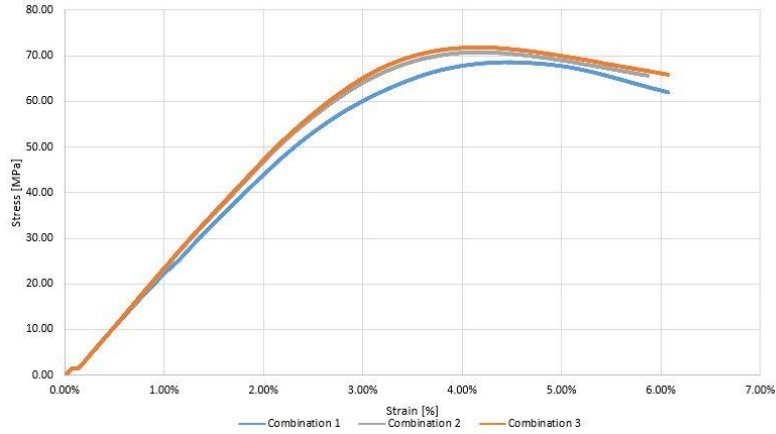


Figure 19 - Optimum combinations of flexural strength.

A maximum stress of 71.90MPa can be observed, which is quite similar to the best tests performed previously.

3.5 Conclusions

After analyzing the results of this study, it is possible to conclude that the maximum stress for the tensile test is 49.96 MPa and for the bending is 72.94 MPa with a standard deviation of 0.89 and 2.09 respectively. It can be admitted right away that flax fiber reinforced composites have higher flexural strength than tensile strength.

With the help of the Taguchi method, it was possible to determine the optimal combinations for maximum tensile and flexural strength. For tensile the optimum combination is A1/A2, B2, C1 and D3, i.e. with/without chemical treatment, with a temperature of 200°C, 10 fibers and a filler percentage of 100%. For bending the ideal combination is A1, B1, C1 and D3, i.e., with chemical treatment, a temperature of 190°C, 10 fibers and a filling percentage of 100%.

With the analysis of variance, it was found that the percentage of filler was the parameter with the greatest contribution to the increase in tensile and flexural strength with percentages of 96.69% and 95.42%, respectively.

After performing the confirmation tests with the ideal combinations obtained previously, maximum values for tensile strength of 48.43MPa and for flexural strength of 71.90MPa were obtained.

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Chapter 4

Tensile and flexural strength of coconut fiber reinforced PLA composites¹

Abstract

Coconut fibers reinforced with PLA composites were analyzed for their tensile and flexural strength characteristics. A Taguchi L18 matrix was created and for each test 4 specimens were run, varying the temperature, the percentage of fibers, the percentage of filler in the specimens, and the arrangement of coconut fibers along the specimen. Regarding the orientation of the coconut fibers, they were placed horizontally and in a net shape. The nozzle temperature used are 190°C, 200°C and 220°C. The percentage of fiber varies by 2, 4 and 6 fiber strands in the case of horizontal orientation and varies by 10, 18 and 34 in the case of cross/network orientation. The filling percentage of the specimen varies between 25%, 50% and 100%. After this, a study of the tensile and flexural strength of the reinforced composites was performed. The optimal combinations for increased tensile and flexural strength were obtained with the parameters for tensile with horizontal fiber orientation, with a nozzle temperature of 200°C, 4 fibers and a filler percentage of 100%. For bending the parameters are with horizontal fiber orientation, a nozzle temperature of 220°C, 2 fibers, and a fill percentage of 100%. With the confirmatory tests a value for the maximum tensile stress of 49.49 MPa was obtained, and for bending it is 85.82 MPa.

Keywords: natural fiber; tensile; flexural; coconut; PLA; additive manufacturing; composite.

¹ Paulo, A. et al., 2022. Tensile and flexural strength of coconut fiber reinforced PLA composites. Journal of Composites Science, it will be submitted in next days.

4.1 Introduction

The best-known first application of natural fiber composites was the use of straw-reinforced clay for building houses, containers and ceramics [1]. The strength of polymers can be improved with the reinforcement of high-strength fibers such as glass fiber, basalt, aramid, among others, thus obtaining the fiber-reinforced polymer composites. Natural fibers come from animal and vegetable fibers, especially flax, hemp, coconut, cotton, sisal, wool, bamboo, etc. [2,3].

Nowadays, more importance is being given to the protection of the environment and consequently incentives have been created for research on agricultural residues. This waste is produced abundantly at low cost and only a small part of this quantity is applied as domestic fuel or fertilizer, and most of it is burned in the field contributing to environmental degradation [4]. An alternative to this problem is the use of this waste as fibers in polymeric materials. These composites have recently been applied in construction, generating properties similar to steel and concrete. These fibers are incorporated into polymer composites because they are low cost, high strength, biodegradable, pose fewer health risks, and have a positive impact on the environment [5,6]. In addition, they produce organic compounds that can be used to produce electricity at the end of their life cycle. Like any other component, they also have disadvantages, such as high moisture absorption, tendency to form aggregates, quality variation and low thermal stability [7,8].

Coconut fiber is extracted from coconut shells, one of the most important natural fillers, from tropical countries such as Malaysia, Indonesia, Thailand, Brazil, and India [9]. Coconut fiber is a lignocellulosic, that is, it consists of lignin and hemicellulose, extracted from the coconut shell [10]. Compared to other fibers, these fibers have less cellulose (36-43%), less hemicellulose content (0.2%), higher lignin content (41-45%), density of 1.25 to 1.5g/m³ and a Young's modulus of 4-6GPa. The lignin content in coconut fiber is quite high, so that the fiber becomes stiffer and stronger [11]. Like all natural fibers, coconut fibers have several characteristics such as low conductivity, low density, low cost, and are lightweight [12]. Several studies and research have been conducted in coconut fiber development for applications in polymer composites [11]. Coconut fibers have a traditional use as yarns, ropes, covering materials and more recently have been applied in acoustic and thermal insulation [13,14].

Composite properties depend on several factors, mainly on fiber volume fraction, orientation, surface quality, hydrophilicity and manufacturing process parameters [15]. The fiber orientation is very important in controlling the mechanical properties of the composite. Several studies point out that by varying the orientation angle between 0° and 90°, one notices several significant changes [16–19].

Biodegradable natural fiber reinforced composites are made of completely biodegradable polymeric matrices, including polylactic acid (PLA) and polybutylene succinate (PBS) [20]. The development of composites with coconut fiber, provides an alternative to composites reinforced with synthetic fibers, and there are several benefits, such as better mechanical and thermal properties, excellent biodegradability, and cost reduction due to the replacement of the biopolymer with cheaper natural fibers. The use of PLA/coconut fiber biocomposites, facilitates the reuse of coconut, thus reducing the environmental impact on landfills [21–23].

There are many polymers from renewable materials such as polylactic acid, polyhydroxyl butyralates, starch-based plastics, and lignin. These have disadvantages such as low commercial availability, low toughness, high price and poor moisture resistance [7]. PLA is very versatile because it is a type of filament for the 3D printer and can be decomposed naturally because it is produced from renewable agricultural raw materials and is obtained by fermenting corn starch into lactic acid, which reduces the environmental impact [7,24]. Polymers are stiff and brittle, so it is often necessary to use plasticizers to improve elongation and impact properties. Its degradation occurs due to hydrolysis into lactic acid, when it is metabolized by microorganisms into water and carbon monoxide. When this polymer is in contact with another biomass, it biodegrades in about 2 weeks, and its material disappears completely in 3 to 4 weeks [25]. Demand for biodegradable polymers is expected to increase substantially in the coming years, indicating that further commercial development of these products is needed [7].

3D printing is defined as a process of creating an object by adding layer upon layer or joining a material based on 3D model data, aided by a computer. It is distinguished from traditional subtractive and equal-material manufacturing methods by the layered accumulation of the material through the printhead, print nozzle, and other printing techniques [26]. The materials printed in 3D, can generate various products from the smallest to the largest, depending on the type of printer that is used [27–29]. Recently, the use of additive manufacturing for natural fiber reinforced polymer composite

materials has gained much interest from educational institutions, leading to several research on processing and molding techniques and mechanical properties of biodegradable composites [30,31].

The objective of this study is to compare the tensile and flexural strength of coconut fiber reinforced composites, analyzing the influence that the amount of coconut fiber, temperature, percentage of specimen filling and fiber orientation have on the mechanical strength of the composites.

4.2 Materials and Methods

In this chapter, the materials and methods used for the tensile and flexural tests will be discussed.

4.2.1. Materials Used

4.2.1.1. Horizontally oriented and cross/networked specimens

To develop these test specimens, it was necessary to use a 750g roll of EasyFil PLA filament of FormFutura brand with 1.75mm, for a printing temperature between 180°C and 220°C. This filament was chosen because it is a biodegradable thermoplastic and has the characteristics of being easy to print, low deformation, excellent adhesion between layers and good impact resistance.

An Anycubic 3D printer was used to manufacture the specimens. Coconut fibers are used in order to reinforce the composites. Using an MDF mold, made in the FabLab, it was possible to maintain coherence between the fibers and with the help of tape and scissors to fix them.

4.2.1.2. Tensile and flexural tests

To perform these tests, it was essential to use previously made specimens, a Shimadzu universal testing machine, varying the type of grips as needed to perform the tensile or bending tests, and a computer to enter the specimen data and collect the data.

4.2.2. Methods

4.2.2.1. Preparation of the test specimens with horizontal orientation

The first step was to glue the fibers in the mold, with tape, according to the number of fiber threads that would be intended, that is, with 2, 4 or 6 threads, fixing them, restricting their movements, since in the mold created are inserted small grooves to place the fixed fibers. When this is finished, the additive manufacturing process begins, placing

the PLA and starting the program that was previously inserted into the 3D printer to form the specimens. At the beginning of the printing process, and as soon as half of the specimen is obtained, the printer is paused, and the mold fibers are placed (Figure 20 (a)). With the help of the tape, the fibers were stretched so that they were fixed, and the printer could not damage them. Printing continues to finish the specimens. After finishing this process, which takes about 1 hour 15 minutes for tensile tests and 2 hours for bending tests, it is necessary to let the machine cool down to remove the specimens. To organize the specimens, they were grouped 4 by 4, for the respective tests and identifying their characteristics (Figure 20 (b)).

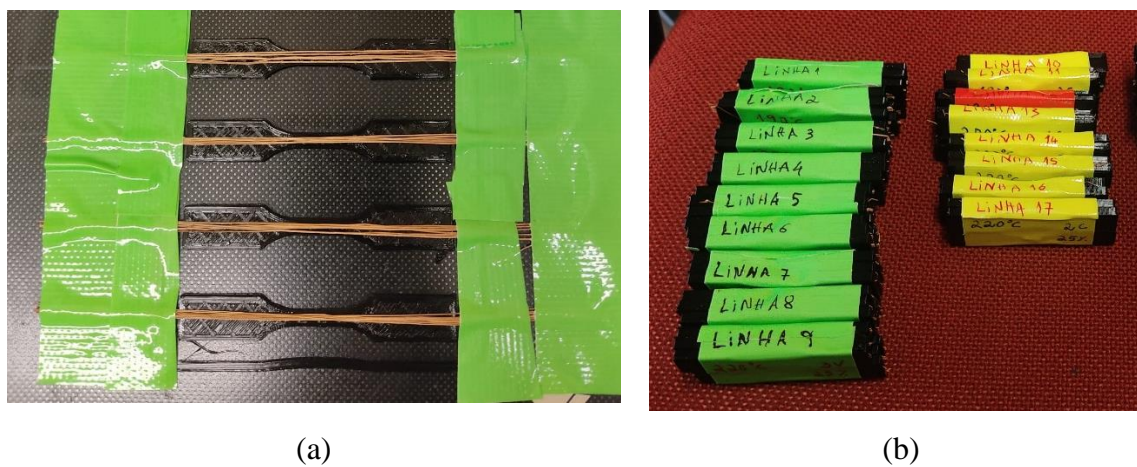


Figure 20 - Preparation of the specimens with horizontal orientation (a) and identification of their characteristics (b).

4.2.2.2. Preparation of cross-oriented/networked specimens

These specimens were made in an identical manner to the specimens with horizontal orientation (Figure 21), differing in the mold in which the fibers were placed, the fiber orientation, the number of fibers used and the time to make the specimens.

Three levels of variation in the number of fiber strands were used. The first level corresponds to 10 threads, the second to 18 threads, and the third to 34 threads. For this process an MDF mold was created with grooves differentiated by levels in order to facilitate the visualization of the number of fibers that were being used and that the angle between fibers was as precise as possible to obtain consistency in the results. The fibers were placed in the mold perpendicularly, in a net shape, i.e. crossed over each other. For the preparation of the specimens, the time was also different because while for the horizontally oriented specimens 4 specimens were made at once, for the cross-oriented/networked specimens only 2 were made due to the space that had to be left to place the fibers. Thus, the tensile specimens took about 38 minutes and the flexure

specimens 1 hour.



Figure 21 - Preparation of cross-oriented specimens.

4. 2.2.3. Preparation of tensile tests

The tensile tests were performed at LERM, i.e., the laboratory of structures and strength of materials. Initially the machine and the computer were turned on and the parameters for the specimen measurements, the test speed, and what kind of results would be desired were entered. The standard that regulates this type of test is ASTM D638.14 [32] using the dimensions 63x10x4mm and a test speed of 10mm/min, and the specimen design can be seen in Figure 22. After that, each specimen was inserted, in turn, in the machine, stipulating the appropriate measure between claws and with the help of the computer we proceeded to the test. Once all tests were finished, the graphs and data obtained were checked and extracted to an Excel file in order to process them.

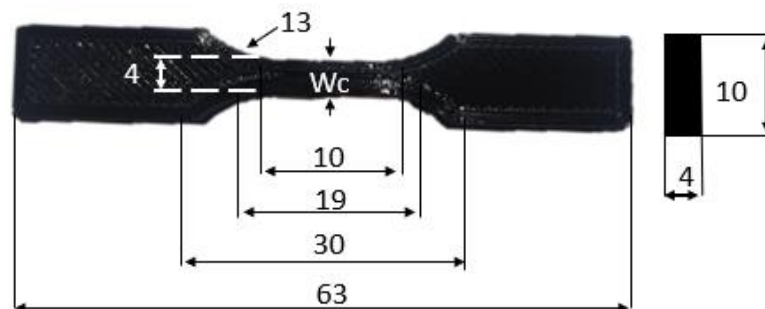


Figure 22 – Specimen dimensions for tensile tests in [mm].

The tensile strength can be calculated from the ultimate load and the cross-sectional area. For the representation of the stress-strain curve it is necessary to determine for each point the respective values. The stress is calculated by dividing the applied load by the average cross-sectional area.

$$\sigma = \frac{P}{A} \quad (5)$$

Where:

σ : Axial tensile stress at one point (MPa)

P : Load applied at the point (N)

A : Average cross sectional area (mm²)

The strain can be calculated by subtracting the initial length from the length, dividing everything by the initial length, i.e.:

$$\varepsilon = \frac{L-L_0}{L_0} \quad (6)$$

Where:

ε : Strain

L : Specimen length (mm)

L_0 : Initial length of specimen (mm)

4.2.2.4. Preparation of the bending tests

These tests were performed in the same laboratory as the tensile tests, in the same way, i.e., the machine and the computer were turned on and the parameters for the specimen measurements, the test speed and what kind of results were desired. The ISO 14125 standard [33] was used, using the measurements 80x10x4mm and a test speed of 1.7mm/min.

The test speed was calculated using the expression:

$$V = \frac{\varepsilon' L^2}{6h} \quad (1)$$

Being:

V : Test speed (mm/min)

ε' : Deformation rate of 1%/min

L : Outer span (mm)

h : Thickness (mm)

After that, each specimen was inserted, in turn, in the machine, stipulating the

appropriate measure between claws and with the help of the computer we proceeded to the test. After finishing all the tests, the graphs and data obtained were checked and extracted to an Excel file in order to process them.

These tests were performed at 3 points (Figure 23), i.e., the specimen is positioned on two parallel supports and is flexed by a cleaver. The specimen used has the shape of a flat beam of constant rectangular cross section. In three-point bending, the maximum bending stress occurs at the external surface of the specimen and is given by:

$$\sigma = \frac{3PL}{2bh^2} \quad (2)$$

Where:

σ : Bending stress on outer surface (MPa)

P : Load applied at the point (N)

L : Specimen length (mm)

b : Specimen width (mm)

h : Specimen Thickness (mm)

The strain is calculated from the formula:

$$\varepsilon = \frac{6\delta h}{L^2} \quad (3)$$

Where:

ε : Strain

δ : Deflection - Distance of the bottom or top surface of the specimen in the middle of the span that has deviated from the initial position (mm)

h : Specimen Thickness (mm)

L : Specimen length (mm)

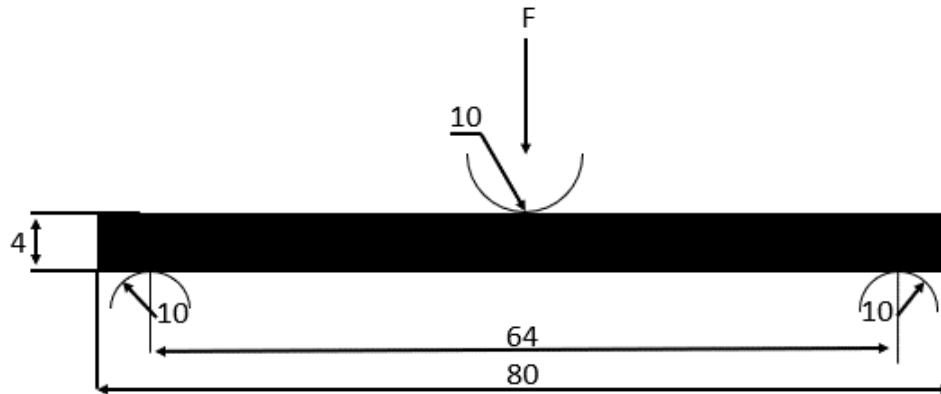


Figure 23 - Specimen dimensions for bending tests in [mm].

In order to simplify the control factors, Table 11 was created for both groups of trials.

Table 11 - Composite control factors.

Symbol	Control factor	Level 1	Level 2	Level 3
A	Fiber orientation	Horizontal	Cross	
B	Nozzle temperature	190°C	200°C	220°C
C	Number of strands	2 or 8	4 or 18	6 or 34
D	Filling level	25%	50%	100%

After the control factors and their levels were defined, it was possible to create a Taguchi L18 array (Table 12). This array was used for the execution of both groups of experimental tests.

Table 12 - Taguchi L18 matrix.

Test number	A	B	C	D
	Fiber orientation	Temperature	Fibers Percentage	Filling level
1	1	1	1	1
2	1	1	2	2
3	1	1	3	3
4	1	2	1	1
5	1	2	2	2
6	1	2	3	3
7	1	3	1	2
8	1	3	2	3
9	1	3	3	1
10	2	1	1	3
11	2	1	2	1
12	2	1	3	2
13	2	2	1	2
14	2	2	2	3
15	2	2	3	1
16	2	3	1	3
17	2	3	2	1
18	2	3	3	2

4.3. Results

This chapter discusses the experimental results obtained by the tensile (Figure 24 (a)) and flexure (Figure 24 (b)) tests, as well as the strength that can be seen in Table 13.

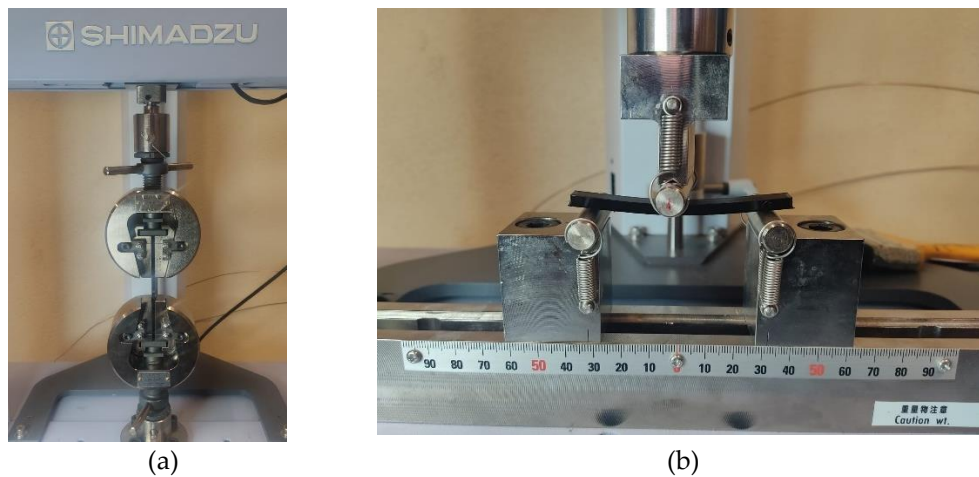


Figure 24 - Tensile tests (a) and Bending tests (b).

After analyzing the results obtained through the universal testing machine, the stress and strain were computed, and a graph was made for each of the 4 specimens of the 18 tests, obtaining a total of 144 specimens for the coconut fiber. Each graph was analyzed, obtaining the following graphs of the averages for each test.

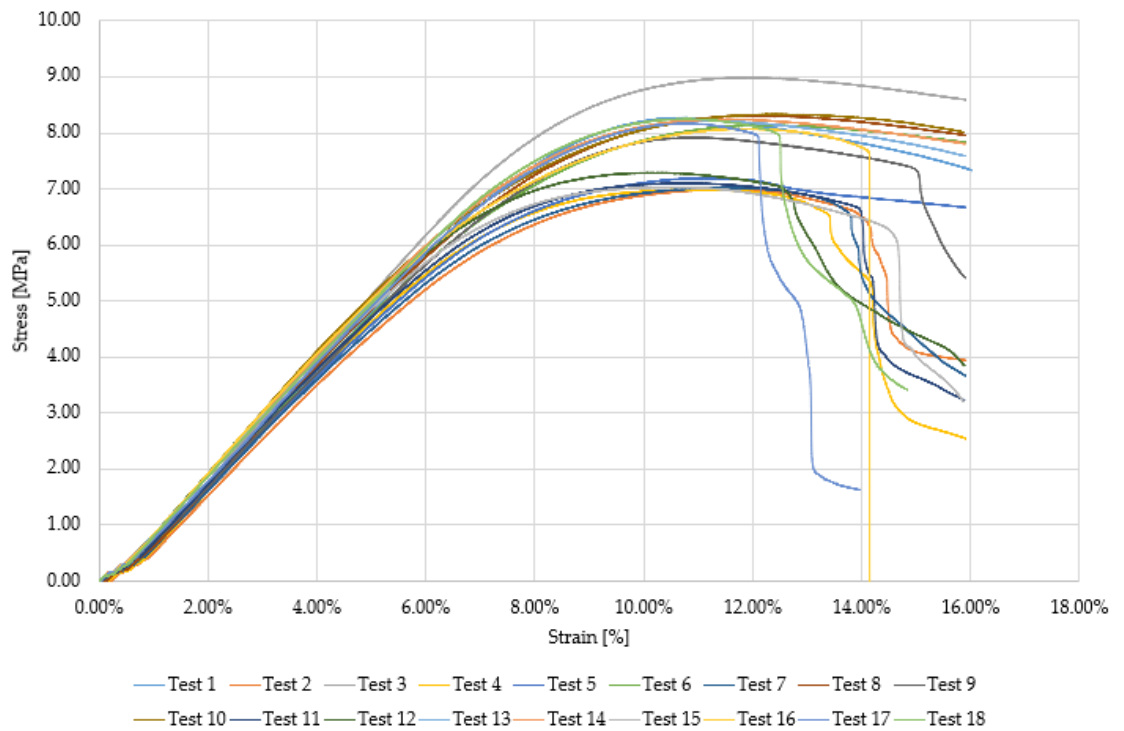


Figure 25 - Stress-strain for tensile strength.

The stress-strain for tensile strength is graphically represented in Figure 25, where it can be seen that test 3 was the one that obtained the best results, followed by tests 8 and 10. Test 3 represents specimens with horizontal fiber orientation at a temperature of 190°C, with 6 fibers and a filling percentage of 100%. Test 8 represents specimens with horizontal fiber orientation at a temperature of 220°C, with 4 fibers and a filling percentage of 100%. Test 10 represents specimens with cross orientation/networked fibers, at a temperature of 190°C, with 10 fibers and a fill percentage of 100%. In other words, what all tests have in common is that the fill percentage is 100%.

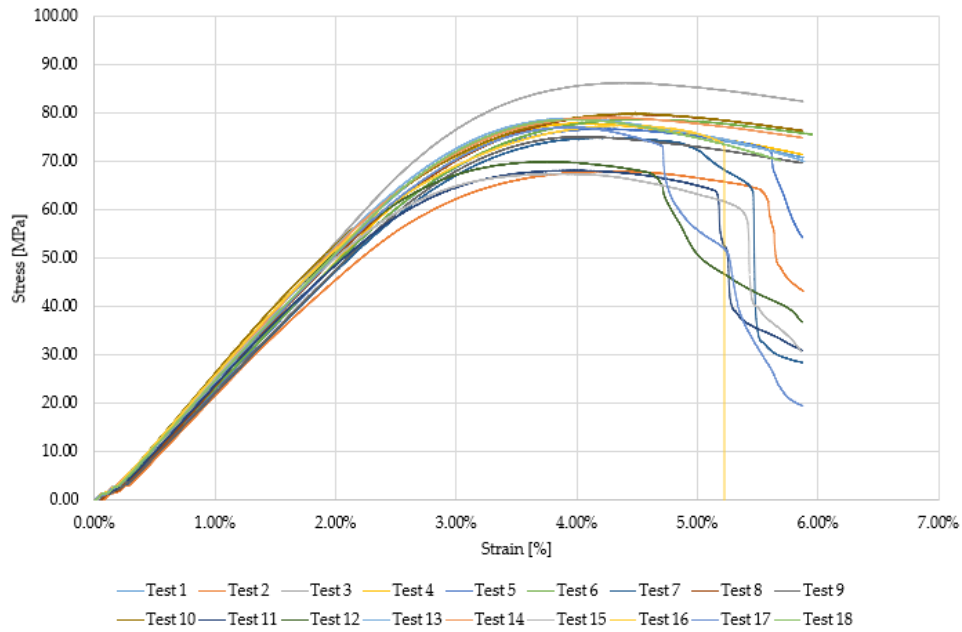


Figure 26 - Stress-strain for flexural strength.

The stress-strain for flexural strength is graphically represented in Figure 26, where it can be seen that test 3 was the one that obtained the best results, followed by tests 8 and 14. Test 3 represents specimens with horizontal fiber orientation at a temperature of 190°C, with 6 fibers and a filling percentage of 100%. Test 8 represents specimens with horizontal fiber orientation at a temperature of 220°C, with 4 fibers and a filling percentage of 100%. Test 14 represents specimens with cross orientation/networked fibers, at a temperature of 200°C, with 18 fibers and a filling percentage of 100%. In other words, what all tests have in common is that the fill percentage is 100%.

Table 13 shows the mean values of the maximum stresses of the 4 specimens in each test, as well as the respective standard deviation. As graphically represented, it can be seen that trial 3 obtained the highest maximum tensile stress value for the tensile test with a value of 49.49MPa and a standard deviation of 0.63 MPa. For the bending tests, the one with the highest stress was also test 3 with a value of 85.82MPa and a standard deviation of 2.51 MPa.

Table 13 - Values of the mean maximum stresses and their standard deviation.

EXPERIMENT NUMBER	TENSIL STRENGTH AVAREAGE [MPa]	DESVIO- PADRÃO [MPa]	FLEXURAL STRENGTH AVAREAGE [MPa]	DESVIO- PADRÃO [MPa]
1	41.26	0.78	74.41	4.78
2	43.04	1.02	70.06	4.32
3	49.49	0.63	85.82	2.51
4	42.00	0.68	72.88	5.19
5	44.57	0.97	73.21	4.20
6	45.21	1.76	78.39	0.79
7	41.81	1.17	73.08	3.24
8	46.55	1.33	79.95	1.58
9	40.08	0.41	72.87	4.09
10	45.82	1.38	78.51	1.50
11	40.95	1.09	68.23	0.97
12	38.77	1.84	66.02	10.82
13	43.88	2.73	77.44	3.16
14	44.76	1.17	78.97	0.70
15	41.48	1.45	67.28	5.92
16	43.91	2.87	78.09	1.89
17	41.18	0.49	75.42	4.70
18	37.76	1.42	75.38	5.82

4.4. Discussion

With the S/N ratio suggested by Taguchi, it is possible to determine the quality characteristics that deviate from the desired values, so it is necessary to convert the experimental results into a Signal-to-Noise ratio (S/N). This ratio measures the performance of the levels in order to choose the control factors that reduce noise. In other words, the noise means the unwanted values (that you want to decrease) and the signal means the output characteristic that you want to achieve. This relationship considers the mean and variance, so it can be assumed that the mean will be the signal and the standard deviation the noise. The characteristic to be optimized will always be the highest S/N ratio criterion. There are 3 categories for the S/N ratio analysis, being the lowest better, the highest better and the nominal best. The factor level for the S/N ratio is calculated using the S/N analysis. The best quality characteristics are always corresponding to a higher S/N ratio, regardless of the category under study. Therefore, the optimal level of control factors is the level with the highest S/N ratio. To determine the statistically feasible control factors it is necessary to use the analysis of variance.

In this study, it is intended to maximize the tensile and flexural strength of coconut fiber-reinforced composites, so the category of quality characteristics for the S/N ratio is the highest-best:

$$S/N = -10 \log \left(\frac{1}{n} \sum_{i=1}^n \frac{1}{y_i^2} \right) \quad (4)$$

where n is the number of observations and y_i is the observed data [34,35].

Table 14 shows the S/N ratios for tensile and flexural strength.

Table 14 - S/N ratios for tensile and flexural strength.

TEST NUMBER	A	B	C	D	S/N_{ts} ratio [dB]	S/N_{fs} ratio [dB]
1	1	1	1	1	32.31	37.38
2	1	1	2	2	32.67	36.86
3	1	1	3	3	33.89	38.66
4	1	2	1	1	32.46	37.19
5	1	2	2	2	32.97	37.25
6	1	2	3	3	33.08	37.88
7	1	3	1	2	32.42	37.25
8	1	3	2	3	33.35	38.05
9	1	3	3	1	32.06	37.21
10	2	1	1	3	33.21	37.89
11	2	1	2	1	32.24	36.68
12	2	1	3	2	31.74	35.95
13	2	2	1	2	32.79	37.76
14	2	2	2	3	33.01	37.95
15	2	2	3	1	32.34	36.46
16	2	3	1	3	32.79	37.84
17	2	3	2	1	32.29	37.49
18	2	3	3	2	31.52	37.46

In Table 14, S/N_{ts} is the S/N ratio for tensile strength and S/N_{fs} is the S/N ratio for flexural strength, where the S/N results for the 18 combinations L18.

For a higher S/N ratio, the best category is applied in order to maximize the response (tensile and flexural strength). The average S/N ratio for the control factors of levels 1, 2 and 3 can be calculated by averaging the S/N ratios of the corresponding tests. In Table 15 and 16, the average S/N ratio for each level of control factor is shown, i.e., the response.

Table 15 - Mean response table of the S/N ratio for tensile strength and significant interaction.

Symbol	Control factor	Mean S/N ratio [dB]		
		Level 1	Level 2	Level 3
A	Fiber orientation	32.80	32.44	
B	Temperature	32.67	32.78	32.41
C	Fibers Percentage	32.66	32.76	32.44
D	Filling level	32.28	32.35	33.22

Table 16 - Mean response table of the S/N ratio for flexural strength and significant interaction.

Symbol	Control factor	Mean S/N ratio [dB]		
		Level 1	Level 2	Level 3
A	Fiber orientation	37.53	37.28	
B	Temperature	37.24	37.41	37.55
C	Fibers Percentage	37.55	37.38	37.27
D	Filling level	37.07	37.09	38.05

Figures 27 and 28 show the response graph of the S/N ratio for tensile and flexural strength, respectively. By analyzing the figures, it is possible to determine the optimal combinations for increasing tensile and flexural strength.

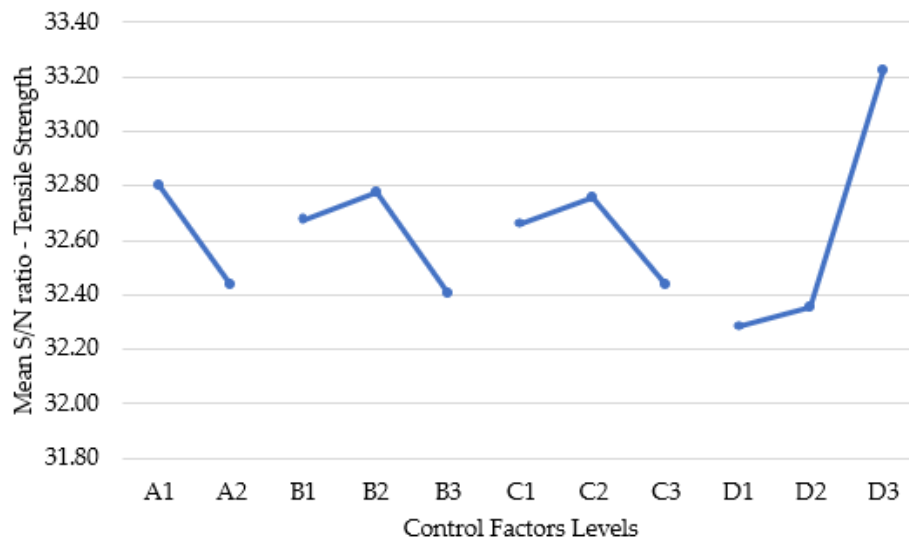


Figure 27 - Response graph of the S/N ratio for tensile strength.

Through the analysis of Figure 27 it is possible to determine the ideal combinations to increase the tensile strength of the coconut fiber reinforced composite. Regarding the tensile tests, the ideal combination is A1, B2, C2 and D3, that is, with horizontal fiber orientation, with a temperature of 200°C, 4 fibers and a filler percentage of 100%.

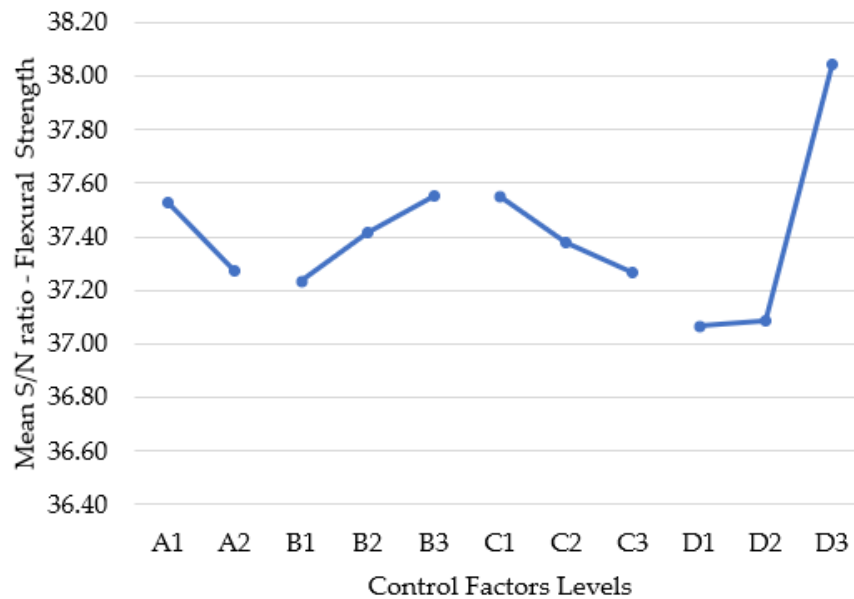


Figure 28 - Response plot of the S/N ratio for flexural strength.

Through the analysis of Figure 28 it is possible to determine the optimal combinations to increase the flexural strength of the coconut fiber reinforced composite. For bending tests, the ideal combination is A1, B3, C1, and D3, i.e., with horizontal fiber orientation, a temperature of 220°C, 2 fibers, and a fill percentage of 100%.

It is necessary to identify more precisely the relative importance of the control factors for tensile and flexural strength, for which ANOVA analysis is used in order to determine the optimal combinations of the levels of the control factors. Through Tables 17 and 18, it is possible to verify what most influences the values of tensile strength and flexural strength, as the percentage of filling of the specimens is the one that obtains a higher percentage, indicates that the more percentage of filling it has the better.

Using Tables 17 and 18, is possible to see the variance results for each control factor, where DF is the degree of Freedom, Adj SS is the sum of squares and Adj MS are the mean squares. The F-test is a statistical tool to check which parameters significantly affect the quality of the characteristics, i.e. it is defined as the ratio of the mean square deviations to the mean square error.

Table 17 - ANOVA for tensile strength.

Source	DF	Adj SS	Adj MS	F-Value	P-Value	Contribution [%]
A	1	0.59	0.59	4.98	0.05	10.18%
B	2	0.44	0.22	1.86	0.21	7.59%
C	2	0.32	0.16	1.33	0.31	5.45%
D	2	3.29	1.64	13.79	0.00	56.34%
Error	10	1.19	0.12			20.43%
Total	17	5.83				100.00%

Table 18 - ANOVA for flexural strength.

Source	DF	Adj SS	Adj MS	F-Value	P-Value	Contribution [%]
A	1	0.28	0.28	1.14	0.31	3.98%
B	2	0.29	0.15	0.60	0.57	4.18%
C	2	0.24	0.12	0.49	0.63	3.43%
D	2	3.77	1.88	7.62	0.01	53.38%
Error	10	2.47	0.25			35.03%
Total	17	7.06				100.00%

After analyzing the results of the F test value, it is possible to verify that for the tensile strength, the most significant control factors are fiber orientation and the filling percentage of the specimen, with the remaining factors varying. For the bending tests, the most significant control factors are temperature and specimen fill percentage, with the remaining factors varying. That is, for tensile strength, the percentage of filler was significant with 56.34%, followed by fiber orientation, nozzle temperature and the percentage of fibers with 10.18%, 7.59% and 5.45%, respectively. For flexural strength, the percentage of filler was also the most significant with 53.38%, followed by the nozzle temperature, fiber orientation and fiber percentage with 4.18%, 3.98% and 3.43%, respectively.

4.4.1. Confirmation Tests

Based on the results, tensile and flexural tests were performed for the best combinations, and 4 specimens were tested for each of the tests. For the tensile tests, the specimens were run with horizontal fiber orientation, with a temperature of 200°C, 4 fibers, and a filling percentage of 100%. The bending tests were done with horizontal fiber orientation, a temperature of 220°C, 2 fibers, and a fill percentage of 100%.

Figure 29 graphically represents the optimal combinations of tensile strength.

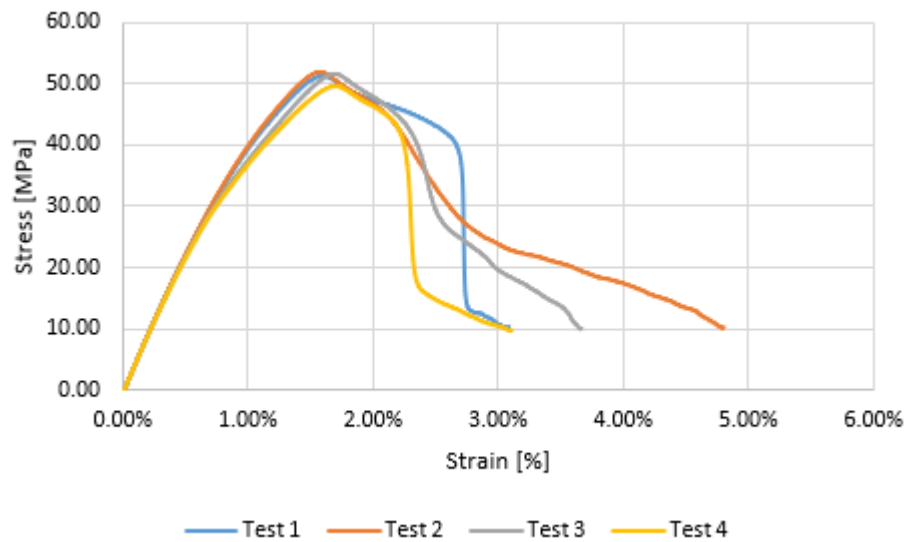


Figure 29 - Optimum combinations of tensile strength.

A maximum stress of 51.99MPa can be observed, which represents a higher result, although quite close to the best tests performed previously.

Figure 30 graphically represents the optimal combinations of flexural strength.

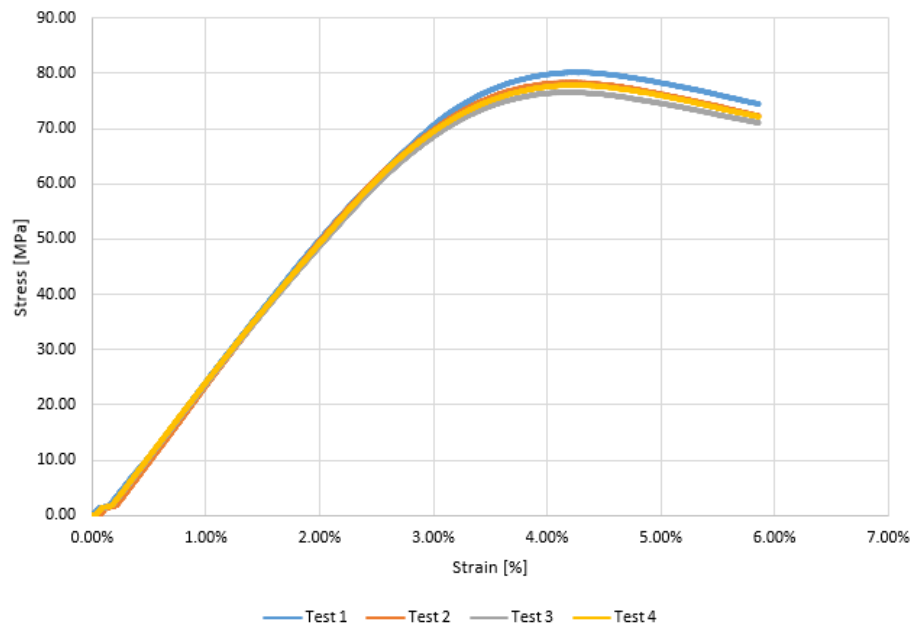


Figure 30 - Optimal combinations of flexural strength.

A maximum stress of 80.19MPa can be observed, i.e., it is a stress quite similar to those of the best tests performed previously.

4.5. Conclusions

Several conclusions can be drawn from this study. One can start by concluding that the flexural strength of the coconut fiber reinforced composite is higher than the tensile strength of this composite, with values of 85.82MPa and 49.49MPa, obtaining a standard deviation of 0.63 and 2.51, respectively.

With the use of the Taguchi array and its result analysis method, it was found that the ideal combinations to increase tensile strength are A1, B2, C2, and D3, and to increase flexural strength are A1, B3, C1, and D3. Thus, for tensile strength a horizontal fiber orientation, with a temperature of 200°C, 4 fibers and a filling percentage of 100% is required, on the other hand, to increase the flexural strength the optimal combination is when using a horizontal fiber orientation, a temperature of 220°C, 2 fibers and a filling percentage of 100%. From these combinations it can be seen that in both cases, the use of fibers in a horizontal orientation and a filling percentage of 100% is preferable.

After performing the confirmation tests with the ideal combinations obtained previously, maximum values for tensile strength of 51.99MPa and for flexural strength of 80.19MPa were obtained.

After studying the results validated by ANOVA analysis, it was found that as expected, the percentage of filler is the parameter with the greatest contribution to increasing the strength of the composites in both tests performed, with a contribution percentage of 56.34% for tensile tests and 53.38% for bending tests.

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Chapter 5

Conclusion and Future Directions

This last chapter will present the final conclusions that can be drawn after analyzing the results of this study. Suggestions for future work to follow up on this study will also be presented.

5.1 Conclusions

During the present study, the feasibility of using flax and coir fibers to reinforce composites was studied. After a literature review it was understood the current need to replace plastics and synthetic fibers, with environmentally friendly materials, hence the need to use a biodegradable composite (PLA) and natural vegetable fibers, flax and coconut.

After the specimens were prepared, tensile and flexural tests were performed.

Considering the results obtained for the flax fiber, it was verified that it presents a higher flexural resistance, with values of 72.94MPa compared to 49.96MPa obtained in the tensile tests.

In this fiber the influence of using an alkaline treatment with NaOH was studied, however, after verifying the ideal combinations, it was verified that this treatment did not have significant influence for the tensile tests, however, in the bending test the use of chemical treatment brought improvements in the composite's resistance. Therefore, the optimal combinations to increase the tensile strength of these composites was A1/A2, B2, C1 and D3, i.e. with/without chemical treatment, with a nozzle temperature of 200°C, 10 fibers and a filler percentage of 100%. On the other hand, in bending tests the ideal combination is A1, B1, C1 and D3, that is, with chemical treatment, a temperature of 190°C, 10 fibers and a filling percentage of 100%.

Since in the flax tests the alkaline treatment did not show the expected results, in the tests with coconut fiber, the influence of this chemical treatment was not studied; on the other hand, a physical alteration was studied, i.e., placing the fibers in a net shape. In this experimental group, the bending tests also presented better results than the tensile tests, with values of 85.82 MPa and 49.49MPa, respectively. In relation to the physical change (fibers in a net) there were worse results, that is, the application of the fibers only

horizontally shows better results. With the optimal combinations for increased tensile strength is A1, B2, C2 and D3, i.e. a horizontal fiber orientation, with a nozzle temperature of 200°C, 4 fibers and a filler percentage of 100%. For improved flexural strength the ideal combination is A1, B3, C1 and D3, i.e. horizontal fiber orientation, a temperature of 220°C, 2 fibers and a filler percentage of 100%.

With the analysis of variance, it was found that in all tests the percentage of filler was the parameter with the greatest influence on increasing the strength of composites to the tests submitted, and in tests with flax fiber showed values between 95.42% and 96.69% and for coconut fibers values in the range of 53.38% and 56.34%.

After performing the confirmation tests with the ideal combinations obtained previously, maximum values for tensile strength of 51.99MPa and for flexural strength of 80.19MPa were obtained.

After performing the confirmation tests with the ideal combinations obtained, maximum values for tensile strength of 48.43MPa and 51.99MPa were obtained for the composites reinforced with flax and coconut fibers respectively. For the flexural strength values of 71.90 MPa were obtained for the flax fiber reinforced composites and 80.19 MPa for the coconut fiber reinforced composites. In conclusion, coconut fibers showed better results, that is, coconut fiber-reinforced composites are more resistant to tensile and flexural strength.

5.2 Future Directions

During this study, important variables related to the strength of materials were studied.

It would be interesting to modify the 3D printer, adding a roller so that when the printer applies the PLA it also adds the fiber, thus the composite would be completely reinforced with fiber and the PLA - Fiber bond would be greater.

As a future project, it may also be interesting to make PLA composites with the junction of the two studied fibers or filament manufacturing with the incorporation of natural fibers.