

Use of vegetable fibers in polymer composites: a review with a view to their application in designing new products

Uso de fibras vegetales en materiales compuestos de matriz polimérica: una revisión con miras a su aplicación en el diseño de nuevos productos

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Abstract

Natural fibers are becoming a really striking alternative for industrial applications because of its low cost, light weight and as a renewable raw material. This type of fibers shows better properties than other conventional materials when used as reinforcement in polymeric composite materials (PCM). Because of these facts, these materials are being used in footwear and leather goods, rubber parts, vehicles and building industry. This work is a review about the use of cotton, fique, jute and hemp in PCM, starting with the classification of vegetable fibers and its properties. The evolution of natural fibers as raw materials for industry and the key challenges for manufacturing articles with high performance properties are described. Remarkable considerations for designers about properties of PCM are stated.

Keywords: polymer composites; natural fiber; biodegradable composites; mechanical strength; applications.

Introduction

During the last decades, use of polymeric materials has meaningfully increased in their most diverse usage, due to the fact that they have important advantages in comparison to other materials, such as, easy processing, the productivity that its collection can achieve and its low cost (Albinante *et al.*, 2013; Meza *et al.*, 2015). It is a common practice to improve the polymeric materials through the incorporation of particles, tissues or fibers of different

natures, obtaining composite materials of polymeric (Albinante et al., 2013; Araujo, et al., 2008; Mukhopadhyay and Fangueiro, 2009), elastomer (Mohanty, 2005; Nair and Joseph, 2014) or thermo-stable matrix (Meza et al., 2015; Raju and Kumarappa, 2012; Shih, 2007; Yousif et al., 2012).

Several studies coincide in pointing the advantages of using natural fibers in polymeric matrix composite loads (Baillie, 2004; Bledzki, et al., 2002; Mohanty et al., 2005; Netravali, 2005; Pickering, 2008), among them, they stand out: that they come from renewable resources, which makes its availability easier; are of low cost, biodegradables, thus, mitigating the environmental

impact, and besides, are light. A characteristic of great interest, is that they present mechanical properties that, in some applications, are comparable with synthetic conventional reinforced materials such as carbon or glass fibers (Kalia et al., 2009; La Mantia and Morreale, 2011; Satyanarayana et al., 2007; Satyanarayana et al., 2009).

The natural fibers are classified according to its vegetable, animal or mineral origin. Likewise, the ones from vegetable origin are classified according to the part of they that they are extracted from (Joshi, 2004), as displayed on Figure 1.

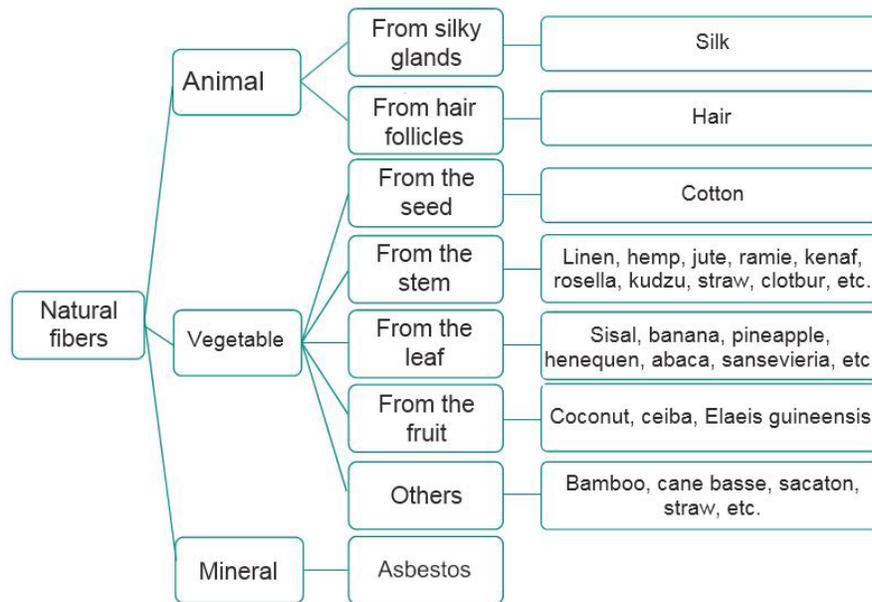


Figure 1. Natural fibers classified according to its origin
Source: (John and Thomas, 2008).

Vegetable fibers used in polymeric matrix composites

The presented revision in this work, addresses the use of vegetable fibers, such as, cotton, fique, hemp and jute, for being the most consumed for the polymeric matrix composites, even acting as enhancers in numerous applications. These fibers are constituted mainly of cellulose, lignin, pectin and hemicellulose Thakur, 2013).

The cellulose is the most important structural component in most of the natural fibers of vegetable origin, presenting itself in form of crystalline microfibrils along the length of the fiber (Satyanarayana et al., 2009). It is resistant to hydrolysis and some oxidizing agents,

but it can partially degrade when exposed to strong acids (Thakur, 2013). The cellulose microfibrils are coated by the hemicellulose, a lineal branched chain structure composed of polysaccharides of lesser molecular weight, that allow the linking of cellulose fibers along with the pectin. The hemicellulose is hydrophilic and can be easily hydrolyzed by diluted acids and bases.

The lignin is a polymer that comes from the linking of several coniferyl alcohols, that offer stiffness to the cellular wall, making it resistant to impact and flexion; it is hydrophobic, resistant to acid hydrolysis, and is soluble in Warm alkali and it's of easy oxidation. The tissue lignification, apart from that, provides higher resistant to organism attacks (John and Thomas,

2008). The pectin is a complex anionic polysaccharide, which gives the fibers flexibility, due to the fact that its structure is highly branched (Summerscales et al., 2010; Thakur, 2013).

The cellulose, lignin, pectin and hemicellulose content in the vegetable fibers influence in their properties, being a very common practice to remove the lignin and pectin to enhance the reinforcing effect from the natural fibers

(Dittenber and GangaRao, 2012). A study regarding linen, jute and sisal fibers, showed that the linen fibers degradation starts at a relatively higher temperature compared to other fibers, which is attributed to its low lignin content (Manfredi et al., 2006). On Chart 1 the density and content reported in the literature for cotton, hemp, jute and linen (Moriana et al., 2014; Fiore et al., 2014).

Chart 1. Cotton, fique, hemp and jute chemical composition and density

Fiber	Cellulose (%p/p)	Hemicellulose(%p/p)	Lignin (%wt)	Ashes (%p/p)	Density (g/cm ³)
Cotton	85 - 90	5.7	----	2	1.5 - 1.6
Hemp	69 - 70	20-22	5.5 - 5.7	5,8	----
Jute	72	13	13	2.5	1.3
Linen	81	16.7 – 20.6	3	----	1.50

Source: (Moriana et al., 2014; Fiore et al., 2014).

The cellular content, the microfibril angle and the cellular dimensions influence in the macroscopic properties in the vegetable fibers (Dittenber and GangaRao, 2012; John and Thomas, 2008; Wong et al., 2010; Fiore et al., 2014). The traction resistance and the Young’s modulus increase with the cell length and with the cellulose content, that is to say, with the polymerization grade of such (Methacanon *et al.*, 2010; John y Thomas, 2008). The jute and hemp traction resistance are reported to be in the range of 400 to 800 N/mm² and 550 to 900 N/mm², respectively, properties associated to their high cellulose content (Sen and Reddy, 2011). The microfibril angle is the limit between the fiber axis and the microfibrils, whose diameter is between 10 and 30 nm, and it meaningfully influences in fiber mechanical properties in a way that the more acute is angle, the higher the resistance and stiffness presented by the fibers, and as the angle increases so does the ductility.

A higher fiber porosity (Wong et al., 2010; Yousif et al., 2012), means a higher humidity absorption which decreases the resistance in the fibers (Methacanon et al., 2010). A higher hemicellulose content also increases the humidity absorption, and besides, accelerates the biodegradation process (Yousif and El-Tayeb, 2009).

Up next, the cotton, fique, hemp and jute evolution is described, subject of this study, until employees came to make reinforcements in polymeric matrix materials.

Natural fibers evolution

Cotton

The cotton was the first textile original from Peru, however, it is grown in all regions with mild tropical regions around the world. The fibers coming from the seeds of the cotton is the most important, since it has been used for at least 5000 (Mwaikambo, 2006). On the XIII the European market for cotton went wider and in the XV Century, Venice, Antwerp and Haarlem were important docks for its marketing, such as the selling and transportation of cotton tissues (Thakur, 2013). Its demand has duplicated since the decade of the eighties in the twentieth century. Nowadays, the biggest producer is China, with the 24% of the world production, followed by the US, with a 19%, and India, with a 13%. Its main application is the clothes and shoe wear manufacturing, nevertheless, it is also used in home products such as curtains, upholstery, mattresses and quilts (Begum and Islam, 2013), among others.

Today, innovative cotton fibers are researched for traditional applications, such as the ones researched by Hashem and his contributors (Hashem et al., 2009), achieving cotton tissues free of wrinkles and with more softness, without decreasing their resistance properties (Hashem et al., 2009; Eryuruk, 2012).

Fique

The fique has its origins in the American tropic, mainly in the Andean regions of Colombia, Venezuela and Ecuador, where the right farming weather conditions prevail for almost the whole year (Dhanabalan and Joshi, 2014). Its botanic name is *Furcraea Macrophylla*, also known with the name of sisal (Gómez and Vázquez, 2012). There are diverse sisal varieties, according to the weather conditions of where the crop comes from and the different morphologic characteristics (Ramakrishna, 2010). Nowadays, the sisal is found in the warmest zones in Mexico, and many parts of Africa, West India and many other tropical countries (Gómez and Vázquez, 2012). With the introduction of horizontal looms in America, the Spanish provided a great technological advance in the knitting of these kind of fiber for the manufacturing of espadrilles and textiles, mainly for the sacks manufacturing. At the end of the XIX Century, the sacks demand and production increased a considerable way due to the beginning of coffee exportations; it was then when the eastern Antioquia developed the sack crafting, cordage, and the espadrilles and saddles elaboration. The demand was so big that it was needed to import sacks made out of jute, what induced the Government to encourage the businesses creation (Gómez and Vázquez, 2012). However, with the invention of the synthetic textile fiber of polypropylene, which was being started to use sacks for products transported through the containers, the fique quilts had seen one more time, affected. Thanks to this, every day, people are looking for new applications with an added value for the fique fiber in the textile, industrial and artisanal areas, such as shoe wear fabric, upholstery, curtains, geotextiles, etc. (Gómez and Vázquez, 2012; Gañán and Mondragón, 2002).

Hemp

The hemp is one of the oldest textile fibers, even, struggling with the linen in the XIX century as the main textile fiber of vegetable origin (Small et al., 2002). Due to the fact that it comes from an apiaceous plant, its farming has been forbidden in several countries, but in the last 30 years, meaningful advances have been presented for the legal farming of such on western Europe, especially in the use of this fiber in the manufacturing for clothing and shoe wear fabric, paper and candles, it is also looked for innovative applications such as geotextiles and composite materials (Ranalli and Venturi, 2004).

Jute

The jute is an agricultural product of great importance and one of the most common natural fibers in the third world countries, such as India, China, Bangladesh, etc. The jute industry has an especial importance in the Indian economy and keeps being an important traditional currency source (Dey, 2005; Roul, 2009), however, same as the fique, it faces a harsh competitions in the decoration and furniture competition. It has diverse applications in decoration and furniture materials, such as lamp screens, wall covers, curtains, upholstery, etc. Nowadays, the jute is between the cheapest fibers in the market, since its price is way lesser than to other fibers, such as the glass; and this is why that in terms of volume, it is now the second most important fiber, behind cotton (Mohanty et al., 2005). Currently, one of the seen applications has been given as a reinforcement in composite materials. Although, it possesses some issues, which is why it requires chemical modification in order to improve its humectation for the purpose of improving the fiber-matrix interaction. Rana and Jayachandran (2000), also mention the use of compatibilizing for the improvement of the interaction with these materials.

Taking in account this perspective, several studies have been held with a great variety of natural fibers such as linen, hemp, jute, sisal, coconut fiber, banana, fique, hay, and among many others (Netravali, 2005), at the same time using a wide variety of polymeric matrixes in which the polythene (Albinante et al., 2013), polypropylene (La Mantia y Morreale, 2007, Khalil et al., 2000), polystyrene (Khalil et al., 2000), polyester resins (Lovely, 2004) and natural rubber stand out (La Mantia and Morreale, 2011). Despite its advantages, the use of natural fibers as a material reinforce also cover various challenges. Before describing them, we will talk about the characteristics of the composite materials.

Polymeric matrix with natural fibers

The composite materials are formed by at least one matrix and a natural reinforce of different nature, which keep their physical, chemical and mechanical properties in an individual way, and also, when using the right way we can reach a material that mixes their individual qualities (Moriani et al., 2014). Nowadays, the most of the

composite materials used in the industry are based in polymeric matrixes (Tajet al., 2007).

In India there is a large tradition in the composite production that use natural fibers for the application in pipes, panels and profiles. However, the root of the critics in the last years around the resources preservation, the use of the natural fibers has been re-invented in North America and Western Europe, since these countries have put high interests in the renewable raw materials.

The reinforced polymeric materials with natural fibers, are composites which the matrixes provide them high resistance and stiffness, acting as reinforcing loads that act along with mechanical elements supporting load; the fiber lining and position, apart from that, protect the composite from the environment and possible damages (Begum et al., 2013). It is possible to improve the mechanical properties of the conventional use polymeric materials, through the incorporation of high resistance fibers, widening the application field from the shoe wear sector up to the aero spatial industry, depending on the type of fibers used as a reinforcement and the initial resistance of the starting polymeric matrix.

The main related challenge is the use of natural fibers as a reinforcement material for the polymeric matrix, is the incompatibility between the fibers in the delivering state and the polymeric matrixes, which takes to the formation of aggregates during the processing of the composite and decreasing their durability, its humidity and fire resistance, limit their processing temperatures and produce high variability properties (Dittenber and GangaRao, 2012). The incompatibility is due to the presence of hydroxyl groups in the natural fibers, which turn them hydrophilic, when the polymeric matrixes are hydrophobic (Dittenber and GangaRao, 2012; Xie et al., 2010), leading to a lesser interfacial strength when the glass or carbon fibers are used (Dittenber and GangaRao, 2012). Another disadvantage within the natural fibers, is its low thermal degradation in oxidative environments, lower to 200°C, taking in account that the thermoplastics processing is done with temperatures higher to 200°C, as it has been reported for the natural fibers used in a high density polythene reinforcement (Araújo et al., 2008); however, this behavior is not a limiting to reinforce elastomeric matrixes or a wide range of thermo stable polymers.

In order to improve the compatibility within the natural fibers and the polymeric matrixes, superficial physical and chemical treatments are applied (Dittenber and GangaRao, 2012). The physic methods are applied to

separate the individual fibers in the filaments or alter the fiber morphology, generating pores or higher roughness, and thus, improving the mechanical anchoring with the matrix. In the reported methods, the vapor explosion that saturate the fibers and open their constituting filaments (Mukhopadhyay and Fanguero, 2009), and thermo mechanical methods such as the stretching or calendaring stand out (Bledzki et al., 2002), and also the extrusion, calendaring, molding by compression and molding by injection (Nair and Josep, 2014).

The mechanical treatments look to reduce the fibers' polarity to turn them into less hydrophilic; the most used are the mercerization, acetylation or propionylation and silanization (Albinante et al., 2013). In the mercerization, the fibers are exposed to a sodium hydroxide solution, increasing the superficial roughness in the fiber, which improves the mechanical adherence. The acetylation and propionylation consist in generating an esterification reaction in the hydroxyl group within the fibers, for which these are treated with acetic anhydride or propionic. In the silanization, silanes are used, whose molecules present a terminal hydrophobic group in of the edges, which can develop a van der Waals interaction with the composite matrix, and in the other edge a hydrophilic group that can react with the OH groups of the fiber, thus forming a bridge between the fiber and the matrix; this kind of treatment has been reported for the epoxy resins reinforced with bamboo fibers and particles (Shih et al., 2007), vinyl resins reinforced with bamboo (Chen et al., 2009), polilactic acid reinforced with pineapple leaves (Shih et al., 2014), and epoxy resins reinforced with shredded, sifted and chemically modified peanut shells (Raju and Kumarappa, 2012).

Cotton, fique, hemp and jute applications in composite materials for shoe wear and construction

Applications in the shoe wear industry

The application of natural fibers in the shoe wear industry, is, along with the textile industry, one of the oldest. In this industry parts like suede and insoles have been produced with hemp in polyurethane matrixes (Connor, 1999), incorporating hemp to recycled polyester (Planet Shoes, 2015), or with cork and jute in a natural latex matrix (Rieker, 2015). For the uppers that must be highly flexible, hemp fibers have been implemented to a natural latex rubber matrix (Beltrán, 2011).

An insert of disposable cushion for shoes in order to reduce the forces received by the shoes during the walking and running activity has been patented. The insertion cushion has a first layer of fibrous material composed of cotton, jute, hemp and bamboo, and a second foamed silicon layer joined with the first layer by a polymeric adhesive. The fibers are treated chemically to reduce their humidity and improve their adhesive absorption (Kim, 2012).

Applications in the automotive industry

Nowadays, there are notable tendencies towards the situation of the already established materials by several types of embedded vegetable fibers in the polymeric matrixes. The automotive industry is using linen, hemp, jute, sisal, kenaf or Wood as reinforcement materials. The most used polymeric matrix is the polypropylene, although many other polyurethane applications are emerging (Bledzki et al., 2002), mainly because the natural fibers are lighter than those of glass or carbon.

Fátima and partners studied the acoustic, flammability and biodegradability properties in the low and high density jute, and of the natural reinforced latex rubber composites, for the application in the automotive and architectonic sector. The acoustic properties were measured in terms of normal specific acoustic absorption and the loss of sound transmissions, while the flammability was through the density of the generated fume. The results show that the low density jute is a better absorbent than the high density sound absorbents; also that the natural latex and jute composites deliver a good acoustic isolation compared to the commercially available products. Both materials also showed a good auto extinction capacity (Fátima and Mohanty, 2011).

La compañía Mercedes Benz ha utilizado yute y algodón desde hace más de 15 años en componentes de interiores de sus vehículos, aprovechado también su capacidad de aislamiento acústico y que no se astillan en caso de impacto directo; incluso en la actualidad realizan investigaciones para reducir la utilización de fibras sintéticas (Tapia et al., 2013). También han desarrollado compuestos con funcionalidad estructural, utilizándolos en los paneles de las puertas, en los pilares de la cabina, partes de los asientos, parachoques y apoyacabezas (Pickering, 2008).

Fiat Chrysler Automobiles applies door panels for the vehicles based in reinforced polypropylene with natural kenaf and hemp fibers (Bledzki et al., 2002). The internal panels of the efficient energy in the Audi A2 are made out of polyurethane reinforced with natural fibers, such as linen, sisal or hemp, getting composites with very low densities in comparison to other materials, and a high dimensional stability (Ashori, 2008).

Application in the construction industry

The high raise of the construction sector has implied the development of thermal and acoustic isolators, constantly looking to reduce noise and make better comfort. Traditionally, expensive isolators which didn't use biodegradable materials were used, such as the glass fibers, thermo stable polymeric foams and filling fabrics, but researches have been held with natural fibers such as coconut in polyester (Zulkifh et al., 2008; Zulkifh et al., 2009), jute in natural rubber latex (Fátima and Mohanty, 2011), pineapples in high density polythene matrixes (Araujo et al., 2008), and hemp in several thermo stable matrixes (Dhakar and Zhang, 2015). The use of these fibers has allowed the materials to be suitable for cheap acoustic, biodegradables and recyclable isolators, and from renewable resources. However, a limiting to take in account when using natural fibers in polymeric matrixes in the construction industry is the exposition to the environment, for which is recommended to apply protective coatings or in functionalizing the fibers (Azwa et al., 2013).

Conclusions

The extensive use of natural fibers and their products is due to their low density, high resistance, specific module and stiffness properties, their non-abrasive nature can be used in high loads until obtaining high levels of filling, and their also their biodegradability. On other side the natural fibers are raw materials coming from renewable and highly available sources, which is why being used in industries such as the automotive, the construction, shoe wear and furniture, although for its adequate use it is necessary to generate more knowledge about their properties in order for the designers can include them in new projects.

Due to the fact that they are natural products, their mechanical and physic properties widely vary.

These properties are determined by the cellulose, lignin, hemicellulose and pectin content, as well as the microfibrils structure. Characteristics that depend in the development of each individual and each plant.

The biggest difficulty for the use of natural fibers in polymeric matrixes, is its polar characteristic, which creates mismatch with the matrix; the superficial treatments, physical and chemical, allow to reduce this problem and at the same time the water absorption.

Under certain preparation conditions, the reinforcing effect in the natural fibers is comparable with the one of other widely use synthetics used in the industry, such as the glass fiber. Yet, some natural fibers present more variation in their mechanical properties than the synthetic fibers, which can be tolerated in diverse applications considering the difficulty to recycle the ones from synthetic origin.

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