

Página de la revista: revistas.uis.edu.co/index.php/revistauisingenierias

Bagasse sugarcane fibers as reinforcement agents for natural composites: description and polymer composite applications Fibras de bagazo de caña de azúcar como agentes de refuerzo para compuestos naturales: descripción y aplicaciones de compuestos de polímeros

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Received: 10 February 2019. Accepted: 24 July 2019. Final version: 9 August 2019.

Abstract

Even some natural resources, considered as waste, can be used for manufacturing a lot of products with enhanced sustainable properties, such as cellulosic bagasse. In this review was performed in order to gather state of the art about natural fibers structure, properties, and applications in polymer composites reinforcement, giving an approach of bagasse sugarcane fibers. The literature was done in different scientific databases; more than 50 papers were analyzed. The vegetable fibers are an extensive and multipurpose group, bagasse sugarcane fibers emerge as a remarkable renewable resource, due to their suitable properties and a large amount of available resources worldwide. Nevertheless, this kind of products require the use of adequacy physical and chemical treatments in order to achieve an adequate proper interaction with polymer matrices, additionally, other characteristics as the geometry and fiber content can influence the performance as reinforcements in composite materials.

Keywords: sugarcane bagasse; state of the art; composite materials.

Resumen

Algunos recursos naturales, considerados como residuos, pueden utilizarse para la fabricación de muchos productos con propiedades sostenibles mejoradas, como el bagazo celulósico. Se realizó una revisión para determinar el estado de la técnica acerca de la estructura, las propiedades y las aplicaciones de las fibras naturales en el refuerzo de compuestos poliméricos, con un enfoque acerca de las fibras de bagazo de caña de azúcar La revisión se realizó en diferentes bases de datos científicas, en más de 50 trabajos. Se obtuvo que, de las fibras vegetales, el bagazo de caña de azúcar emerge como un recurso renovable notable, debido a que posee propiedades ventajosas y existe una gran cantidad de este disponible en todo el mundo, no obstante, requiere tratamientos físicos y químicos adecuados para lograr una buena interacción con las matrices poliméricas. También la geometría y el contenido de fibra pueden influir en el rendimiento como refuerzos en materiales compuestos.

Palabras clave: Bagazo de caña de azúcar, estado del arte, materiales compuestos.

ISSN Printed: 1657 - 4583, ISSN Online: 2145 - 8456, CC BY-ND 4.0 How to cite: G. Díaz-Ramírez, M. Maradei-García, G. Vargas-Linares, "Bagasse sugarcane fibers as reinforcement agents for natural composites: description and polymer composite," *Rev. UIS Ing.*, vol. 18, no. 4, pp. 117-130, 2019. doi: 10.18273/revuin.v18n4-2019011

1. Introduction

Since ancient times there is evidence of the use of natural organic fibers in archeological artifacts suggesting the basic use of these materials. The strength of natural fibers was the main property used in lines, ropes, papers and primitive suspension bridges; many of those artifacts are still in use today. The different civilizations have contributed with new fibers sources, process and applications always taking advantage of its properties. Despite the temporary suspension of natural fibers usage due to the growing interest for plastic materials during the world war I and II, natural fibers rebirth in the 90s as reinforcements applications.[1] Initially, synthetic plastic fibers satisfied the worldwide requirements of versatile end economic materials without an environmental impact evaluation. Nowadays, the consequences of this out of scale development on the environment are evident, this has raised awareness about the importance of the use of sustainable resources like natural fibers.

Natural fibers can be classified as a type of renewable material extracted from a vegetable, animal o mineral sources (Figure 1). Mineral and animal fibers are out of the scope of this work by the cancerogenic and protein nature of this kind of fibers, respectively. In general, *natural fibers* (NF) refer to plant/vegetable fibers more than animal ones, because this kind of materials are the most utilized and applied in different engineering fields. A common feature in all vegetable fibers are composed of cellulose, as their major structural component with a hierarchical structure accompanied by well-oriented microstructures [3]. This homogeneous composition offers the opportunity to interact with other fibers from different sources. However, the best performance as reinforcements are achieved by varieties with higher cellulose content [4].

The interest in NF research field it has come in continuous growth, this is reflected in the number of scientific publications that exist today related to them (~ 8000). The amount of studies published have exponentially increased year after year (Figure 2). The NF study field becomes interesting for the academic on natural fibers and cellulose [5]. Many of these publications cover a broad range of issues such as isolation of cellulose from different raw material sources, the chemical and manufacturing process, and a vast number of applications [6] [7].community due to pioneering researches like the developed by Chanzy and Cavaille in the early the 90's



Figure 1. Classification of natural fibers according to their origin. Font: Prepared by the authors and adapted from



Figure 2. Number of publications on natural (plant /vegetable) fibers+ polymer. Font: Prepared by the authors and adapted from Web Of Science

A breakthrough related to NFs was achieved with the incursion of nanocellulose, a new material with properties never seen to date [8]. Nanocellulose refers to cellulosic fibers or crystals with nanometric scale dimensions (1-100 nm in diameter). The advantage of these nano fibers are related to their low density, high specific strength and stiffness, sustainability, small to medium scale production by biological (bacteria) or chemical (hydrolysis/oxidation) process with reasonable yields, low costs, economic processing, and finally, low emission of toxic in eventual final disposal. Nanocellulose can be obtained by different sources like NFs by means of top-down chemical, enzymatic, physical or bottom-up biological methods. In all cases, pretreatments, process and extraction conditions determine the morphology and properties of the obtained nanomaterial.

The natural fibers are an important source of reinforcement materials for alternative polymer composites applications, generating new materials with improved performance, sustainable production (e.g. recyclable, renewable, compostable), competitive edge (e.g. low price, processability, raw material supply and diversity) and applications fields in expansion. On this matter, polymer matrix materials can be classified into thermosets, thermoplastics and elastomer. All kind of polymers are reinforced with fibers, Thermoplastics like polypropylene (PP), polyethylene (PE), polystyrene (PS) and polyvinyl chloride (PVC) are some examples of this type of polymers reinforced with natural fibers.

In general, composites partially substituted with natural fibers have an advantage due to its low toxicity, emission of effluent, energy consumption and abundance of disposal options. This review aims to provide details of the structure of the natural fibers, the properties and their applications in polymeric matrices, providing an approach; where sugarcane fibers are a novel source for composite conception with tailored properties.

2. Method used

2.1. Search parameters

The identification of pertinent literature was done in three different scientific databases: ISI Web of Knowledge, ScienceDirect and Scopus. Furthermore, the keywords for this literature review were placed in two categories, as shown in table 1. The criterion for inclusion in the study were: studies published between 2002 and 2018, in English language and text related to the natural fibers. Studies with other fibers (mineral and animal) were excluded from the analysis. In this way, papers identified through the above search strategy that met the inclusion criteria were reviewed for inclusion in the analysis.

The search was carried out between January and march of 2018. First, papers were selected based on the title, which must be related whit the aim of this study. Then, relevant researches were reviewed and identified through reading of abstract. Finally, only those papers pertinent were read completely.

3. Natural fibers structure

Table 1. Key words for the literature review.

А	В
Fibers	"Renewable
"Natural fibers"	material"
"Vegetable fibers"	Nanocellulose
"plant fibers"	"Nano fibers"
"cellulosic fibers"	"biological
	methods"
Sugarcane	Recyclable
"Sugarcane fibers"	Compostable
	"Sustainable
	production"
Polymers	"New materials"
"polymer matrices"	"Natural
Thermosets	Composites"
Thermoplastics	-
"non-bio-	
degradable	
polymers"	

Font: Prepared by the authors.

The structure of cellulose fiber can be described as a mixture of rigid cellulose embedded in a soft lignin and hemicellulose matrix (Figure 3)[9]. For this reason, natural fibers are also referred to as cellulosic or lignocellulosic fibers. Natural fibers can be described as composites materials which developed naturally by the

combination of cellulose, lignin, hemicellulose, wax, minerals, different materials and others, forming a remarkable bio-engineering material, which differs from the raw materials that originated them. The main component in vegetable fibers are mainly cellulose, that contains 60 % to 80 % of total weight, arranged in microfibrils which are aligned helically along the length of the fiber [10]. The diameter of elementary fibrils is among 2 and 20 nm. Cellulose is a linear macromolecule (homopolymer), which consists of β -*D*-glucose units, linked together by β -1,4-glycosidic linkages at C1 and C4 position to generate a linear polymer chain with a short repeat unit called cellobiose (C₆H₁₁O₅)₂O [11].

The functional groups of repeated units are responsible for intermolecular and intramolecular hydrogen bonds due the occurrence of six hydroxyl groups per unit. This structural feature gives a hydrophilic character to cellulose macromolecule with a polymerization degree between 7000-15.000 [12]. In this respect, due the high number of hydroxyl groups per unit, the intermolecular cellulose hydrogen bonds control de morphology giving a tightly packed, slender or rod-like crystalline structures [13]. Two regions can be distinguished in cellulose fibers; the portion that has a lower packing density is referred to as amorphous cellulose and the second portion is known as *crystalline cellulose* with a straight, stable supra-molecular arrange, consequence of a high packing density. A high cellulose content and low microfibril angle are desirable properties in a fiber to be used as reinforcement in polymer composites [14]. The other components that interact with cellulose influence the fiber wall cell elasticity modulus. Moreover, Hemicellulose is responsible for biodegradation, moisture absorption, and thermal degradation of the fiber. Lastly, lignin, a key component in fiber structure, acts like a natural binder, filling the spaces in the cell wall between pectin, hemicellulose, and cellulose. Lignin has a 3D highly branched polyaromatic structure provide a strong intramolecular bonding [15].



Figure 3. Hierarchical structure of natural fibers describing the size ratio, composition, and structure of cellulose using sugarcane as natural source. Font: Prepared by the authors.



In general, fibers compositions and structures that can be obtained from cellulose vary greatly. depending on plant species, age, climate, and soil conditions; although a basic compositional ratio of lignin/cellulose is preserved on order to ensure the required rigidity [16]. All native natural fibers have a high length compared to their diameter, this condition is also called *high aspect ratio*, and is responsible for their unique properties compared to bulks materials.

4. Properties of natural fibers

Generally, natural fibers properties are closely related to fiber composition, natural origin, growing conditions,

harvesting time, storage procedures and manufacturing processes, this last seems to be a critical factor that define the resulting material performance on strength, reinforcement and biodegradation tests [17]. As example, strength could be reduced by 15 % over 5 days after optimum harvest time, this is related with moisture loss, on the contrary, manually extracted flax fibers have been found to have strength 20 % higher than those extracted mechanically [18]. The properties of each constituent contribute to the overall properties of the fiber, so different fibers with different composition have intrinsic properties (Figure 4a). Figure 4b shows the change in the Young's modulus according to fiber type, showing a high value in this mechanical property for hemp (45 GPa), kenaf (37,5 GPa) and flax (60 GPa) [19].



Figure 4. (a) Young's modulus of different natural fibers and (b) the differences in fibers composition, respectively. Font: Prepared by the autors and adpated from [3, 20]

The structure-properties relationship is associated with interfacial bonds (covalent or hydrogen bonds) between fiber component in its helical arrange defining the mechanical properties of biocomposites, as well as anisotropy through the entire structure [7] [20] This anisotropy refers to the distribution between amorphous and crystalline regions and diverse molecular orientation in fibers resulting in extraordinary mechanical properties.

In general, along with all-natural fibers, the higher variability of mechanical properties is common always in the same fiber direction, but weak and flexible perpendicular to it [22]. Likewise, other physical properties such as electrical or thermal conductivity may be totally different when measured along or perpendicular to the fiber; this aspect open the opportunity to develop tailored composites just controlling of fibers placement (parallel, perpendicular, angulated o mixed). The angle of the microfibrils with respect to the fiber axis has a major role in the fiber properties, given that smaller angles give high strength and stiffness, whereas larger angles provide ductility [23]. Additionally, to promote a mechanical failure requires a large amount of energy to uncoil the spirally oriented microfibrils; in terms of specific stiffness and specific strength natural fibers and glass fibers have similar performance. An insight in fibers qualities shows that the cellulose content and spiral angle is differed from fiber to fiber, even in the same type with a single design [24].

The high surface area is another property that put this materials in the spotlight of many technological applications, mostly when filler or impregnation process are developed on nano fibers [25]. There are several physical properties like fiber dimensions, defects, strength, variability, and crystallinity that are important to know and measure for each natural fiber to reach its highest potential (Figure 5). Moreover, intricate topography and chemistry surface make the natural fibers

a novel platform to increase properties supporting failure strain, tensile strength and stiffness/Young's modulus. Surface modification over fibers can help to improve some properties. Physical (corona, plasma) and chemical (silane, alkaline, acetylation and enzyme) treatment are some of the common modifications; in some cases, the treatment is an essential prerequisite to enable fiber usage [26].

5. Natural fibers and polymeric matrices: composites conception

The manufacturing of composite materials started with ancient but advanced civilizations like Chinese and Egyptian. Natural fibers like flax were used to reinforced clay around 3000 years ago along the banks of the Nile river and its delta. Back in our time, in the '90s were clear that the interest for NFs use has resurfaced once again. Scientists and engineers focus their attention in the extraction, obtaining and study of natural fibers and their composites [27], based on the fact that vegetable fibers can be considered as natural occurring composites, an excellent example for bio-inspirited approaches.

Composites materials are made mainly of two different materials well known as matrix and reinforcement. In composites, matrix performs essential functions such as to protect the reinforcement from the environment, ensure a homogeneous distribution of applied load stress to the reinforcement, and provide a stable shape of the final composite. On the other hand, the reinforcement, were natural fibers appears as a constituent, performs a structural function increasing the matrix properties [28]. The reinforcing effect in composites associated to vegetable fiber is related to the nature of cellulose and its crystallinity A high cellulose content and low microfibril angle are desirable properties in fiber to be used as reinforcement in polymer composites that require high mechanical performance.



Figure 5. Aspects, factors and dependency of natural fibers with properties. Font: Prepared by the authors.



Furthermore, fiber alignment in matrix composite has a strong influence in final mechanical properties. As an example, the highest tensile strength in a NF is achieved at approximately 73 % fiber aligned in composites, imitating the original arrange of native fiber [29]. In more randomly aligned/shorter fiber composites is observed a decrease in strength, presumably due to the higher compaction limit with less aligned fibers. Another aspect about the performance of composites reinforced by natural fiber is the fiber length; at higher fiber length, the mechanical behavior is better. However, exists a critical fiber length for each polymeric matrix, the measure of this critical value helps to understand the mechanical properties of fiber-reinforced composites [30].

In composite materials, reinforcements can be classified according to fiber disposition (Figure 6). The matrix works as a continuous phase surrounding the reinforcement, while fibers are referred to as the discontinuous phase. This phase is generally harder and exhibits mechanical properties superior to those of the continuous phases [31]. Despite matrices can be made of metallic or ceramic materials, there is a wide range of composite applications refers to polymeric matrices. Polymer composites were developed to meet the need for lightweight, high stiffness materials that exhibit additional functionalities, such as wear resistance, electrical properties, and thermal stability. The matrix selection plays a prominent role due to fiber-polymeric matrix adhesion, which determines the mechanical, dynamic and rheological behavior since the stress transfer occurs at the interface matrix-fiber [32]. In composites with low fiber-polymer adhesion were observed that yielding region occurs extending over a large portion of the strain range; in other words, the yielding occurs across low adhesion regions. The main polymer matrices used in composites are PE, PP, PS in thermoplastics; polyester, epoxy and phenolic resins in thermosets polymers and natural rubber in elastomers. PS-jute, PS-sisal, PS-banana/cotton, PS-straw, PSsugarcane, and PS-kapok are some of the promising systems [33]. The use of biopolymers as rubber, chitin, chitosan, and poly (lactic acid) are some of the new trends called green composites, where these biopolymers are used as matrix reinforced with natural fibers. These

fully green composites are environmentally friendly, completely degradable and sustainable, that is, they are truly '*green*' following the green chemistry principles. After use, they can be easily disposed-off or composted without harming the environment [15]. In conclusion, compared to glass or carbon fibers (the most common materials used in reinforcement process) natural fibers benefit from lower density, less tool wear during machining, no health hazards, biodegradability, availability of natural and renewable sources, and lower cost per unit volume basis.

6. Sugarcane – a novel platform

The applications of natural fiber composites are growing in many sectors such as automobile, aerospace and electronics industries, furniture, packing, construction and infrastructure due to the ability to design promising materials with moderate strength, lower cost, and environmental-friendly features [34]. The different kinds of natural fibers such as jute, hemp, kenaf, flax, bamboo and sugarcane that reinforce polymer composite have received great attention. In this group, sugarcane fibers take a prominent place in many applications due to the massive worldwide production (Figure 7). Sugarcane is one of the most abundant crops in many countries, being their main feedstock of sugar and ethanol production [35]. Sugarcane bagasse is a fibrous remaining residue (up to ~ 223 million tons/year) of cane stalks after the crushing and extraction of the juice in the sugar production industry. It is a residue in the trend towards the production of value-added goods from agroindustrial by products [36].

The massive and global production, low cost, low density and substantial mechanical properties of sugarcane bagasse fiber make it an ideal candidate to be considered as reinforcement material in polymer composites. Different researchers have proposed a set of requirements for natural fiber usage in composites such as high cellulose content (> 50%), high crystallinity (> 47%), low metal content (< 5%), tensile strength (170–290 MPa) and modulus of elasticity (15–19 GPa).[37]



Figure 6. Composite classification according to fibers alignment. Font: Prepared by the authors and adapated from [2].

Literature reported values close to 27.1 GPa for Young's modulus with an ultimate tensile strength of 222 MPa and elongation at break of 1.1% for sugarcane bagasse composites, showing their extraordinary reinforcement effect.[38] Nevertheless, a disadvantage of sugarcane bagasse usage resides in their high moisture sorption and weak interface interaction with a polymer matrix. Moisture sorption can be modified performing a hydrophobic treatment on the fiber surface. Interfacial interaction is a common problem associated with cellulose structure that can be improved through surface modifications with chemical coupling agents or compatibilizers that *link* fibers surface with functional groups of the polymer matrix.[39]

6.1. Sugarcane fibers: processing and modifications

Continuous development in design and production and evaluation of natural fiber composite have generated the need for previous processing steps before composite manufacturing. These modifications seek to improve mechanical properties, reductions of structural defects ensuring proper durability, reliability and cost reduction [35]. In general, the processing technics applied for neat polymer are also applied in polymer composites. They involve basic steps such as the impregnation of the fiber with the resin, forming of the structure, curing for thermoset matrices or thermal processing for thermoplastic matrices. This processing techniques include: resing transfer molding, injection molding, extrusion, pultrusion, compression molding [40], filament winding and prepreg textiles among others. Depending on the type of composite, reinforcement size, final application, cost, quantity and quality, a suitable technique can be selected; however, compression molding is one of the most commonly used natural fiber

composite processing technique because it may be suitable for most kind of fibers [41].

Several searchers have suggested specific aspects that can be tailored through fiber processing, they include fiber content, orientation, volume, and diameter [42]. Physical (mechanical activation), nanostructures coating [43] [44] and chemical (acid, alkali or coupling agent) treatments are another kind of approach in sugarcane fibers modifications. Fiber mechanical activation treatment involves the use of high friction, collision, shear and other mechanical actions to change the crystalline structure and, as a result, fiber properties. Sugarcane bagasse fibers of 0.25-0.38 mm average diameter can be obtained using high-energy milling process, this mechanical activation is simple and environmentally friendly compared to chemical methods, as it does not use any chemical solvent [45]. In many cases, the mechanical activation can promote the distortion of chemical bonds resulting in active radicals and functional groups more disposed (in chemical terms) to interact in the polymer matrix interphase. In chemical processing, several methods are available for natural fibers. Alkali (NaOH) treatment was studied by Ridzuan et al. found a reduction in moisture content as well as the diameter of the fiber; this modification (molecular weight reduction) could be associated with the loss of residual water molecules through hydrolysis reaction over fiber micropore [46]. Recent investigations attributed the deterioration of mechanical properties (tensile and flexural properties) with the loss of molecular weight [47]. Acid (HCl) treatment was performed finding in all cases better properties in comparison with unmodified sugarcane bagasse but lower than alkali (NaOH) treatment [48].



Figure 7. Worlwide production of sugarcane and others natural fibers sources. Font: Prepared by the authors and adapted from [23].



An important aspect must be considered in processing and modifications performed over natural fibers in general, the result of this treatments may alter the microstructural re-distribution of reinforcing particles in matrix and the mechanical behavior of composites as well [49]. For the previous reason, small microstructural changes enhance aspects like wettability, surface roughness, viscosity and shrinkage mold reduction in the matrix/fiber composite interphase providing an excellent interfacial adhesion bonding between the matrix and fiber. Careful process modifications on the fiber can enhance above mentioned properties avoiding particle agglomerates that unchain defects or stress concentration sites for crack nucleation and material failure. Overall, alkali can be selected as a primary treatment for all type of natural fibers before any other modification [50].

6.2. SB applications as reinforcement material for composites

Several processes and publications have been reported describing the use of sugarcane bagasse as a raw material for reinforcing composites. Polymer composites based on thermoset resins [51]-[54], foams [55], rubbers [56] and thermoplastic ones such as PE [45], LDPE [57], HDPE [58], PU [59], PLA [60], PP [61] [62], and PVC [63]. Many industries apply natural fiber composite materials in their manufacturing processes, civil engineering [64], construction [65], automotive [66], military [67], aerospace, marine, clothing [68], among others [69], [70]. The main reasons for using the SB as reinforcement polymer composites are the reduction of in environmental impact and import expenses of some products for agricultural production. However, one of the first applications for SB was the use of ashes waste as a replacement for inorganic constituents in the fabrication of tiles for the construction industry. Although the use is economically viable [71], it is disadvantageous because of the high carbon footprint.

There are a lot of studies about alternative uses for SB, in polymer and polymer composites applications. A study carried out for the Cuban institute for sugarcane derivates researches proposed the use of PE recycled reinforced with SB for the packaging of agricultural products.[58] As a result of the research, a polymer composite suitable for injection molding process was developed. This manufacturing process proved to be suitable for SB/thermoset manufacturing and performed under vacuum contribute to improve mechanical composite properties. Additionally, this process allows the fabrication of composites with a homogeneous distribution of sugarcane fibers. Moreover, SB is a proper source of cellulose and nanocellulose for all types of polymer composites[72], due to the crystallinity of cellulose obtained from it. Respect to this, works had been carried out in other areas of high production of sugarcane like Cauca Valley in Colombia [73], where studies demonstrate the need to establish alternatives to the use of sugarcane bagasse, instead of energy production; [74] An alternative mentioned is the use of sugarcane bagasse as a reinforcement in the production of plastic-based mortars. [75]

The main objective in almost all studies is the hydrophilic behavior control of sugarcane fibers, which is a common drawback present in every NF. Composite phases did not have high enough adhesion between the sugarcane fiber and polymer matrices, that can be noticed in their limited mechanical properties. For those cases, a superficial modification must be performed on crude bagasse before to composite manufacturing. This chemical modification encourages the matrix- fiber adhesion reducing the hydrophobic character of fibers[72], in order to make them suitable for polymer composites. Another aspect that motivates the use of SB is the reduction of environmental impact caused by the use of thermoset matrices, this has motivated the use of biopolymer matrices like polyhydroxy butyrate (PHB).[76] On other hand, an alternative application for the SB as a reinforcement agent is in cementitious composites. Several researches proposed the use of SB as a reinforcement agent for concrete [77], [78]. It was found that the use of 1% sugarcane bagasse fibers can be used as lightening because the composite obtained had the same mechanical properties at compressive loads. Similar behavior was founded in other natural sources of fibers like coconut and bamboo.[79] In the construction industry, there are other possibilities for using SB, for example in flooring [71] and insulation boards material fabrication[80].



Sugarcane bagasse has become a viable reinforcement for polymeric composite materials. In this review, several efforts and developments performed to obtain better or unique properties, friendly environmental and a low-cost process for natural fibers composites reinforced with SB were registered, some of them in the Colombian context. This is how, it has been reported the use of SB as an alternative polymer composites reinforcement. improving mechanical properties like tensile resistance, bending and impact resistance until 16% compared with polymers without this type of reinforcement [79]. Several chemical modifications and surface treatments have been recently developed in order to improve the adherence behavior between the raw SB and the polymeric matrices. New processing techniques are available for using these fibers, which allows to position it as a promissory raw material in many industrial applications. There are examples of SB as a reinforcement for thermosets and thermoplastics matrices suitable for injection, extrusion, compression molding and vacuum transfer processes. In Colombia sugar cane is expected to reach 2,318 MT in 2016/2017, concentring the production across the Cauca river with significant contributions of Meta and Santander departments. This massive production involves a proportional sugarcane waste production; this fact constitutes an opportunity for using this material. The challenge is to develop a convenient and environmentally approach that can effectively convert the huge amount of disposable fibers waste into useful appropriate and commercial materials to devise new composites applications.

8. Acknoledgements

German A. Díaz-Ramírez thanks to Colciencias for the Ph D. scholarship 617-2/2013.

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