

The potential of natural fibres for automotive sector - *review* -

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Abstract - Due to environment and sustainability regulation in the last decades considerable performance in green technology in the field of materials science through the development of biocomposites could be noticed. Thus, the development of high-performance materials produced from natural resources expands worldwide. This can be attributed mainly due to their assets compared to their synthetic counterparts like low cost, low weight, less damage to processing equipment, improved surface finish of moulded parts composite, good mechanical properties, biodegradability, abundant and renewable resources. Natural fibres are valuable and versatile resources with multiple advantages.

Nowadays all the more, the automotive industry is under increasing pressure to fulfill environmental and performance demands and higher fuel efficiency at competitive costs. Automakers recognize potential in biocomposites if these materials can offer the same performance as traditional composites but with lower weight. Additionally they exhibit non-brittle fracture on impact, which is another significant requirement for automotive sector. Other drivers that scores for use of natural fibres reinforced polymer composites (NFRPC) in automotive applications imply reduced waste disposal, reduction of greenhouse gas emission and Life Cycle Consideration.

In spite of their benefits, the significant challenge for producers and supplier to handle with natural fiber reinforced polymer composites resides in large inconsistency of their properties. The chemical composition of vegetal fibres relies on several factors comprising fiber variety, time of harvesting, climatic history, soil characteristics and fibre processing technology. All these factors exert an influence on their final properties when used as reinforcements in biocomposite materials.

In this review a wide range of issues is addressed with special reference to mechanical properties of fibres, interface adhesion and environmental implication of NFRPC. The discussion on the cellulosic/lignocellulosic fibre properties is conducted in order to relate their chemical composition, microstructure and mechanical properties and to understand their *use* and *limits* as reinforcements in composite materials.

The variation within the mechanical properties of natural fibres is a challenge towards designing predictable components for industry since the engineers are accustomed to the precise and reproducible properties of synthetic fibres.

The hydrophilic nature of lignocellulosic fibres causes poor resistance to moisture and incompatibility to hydrophobic polymer matrix. As a consequence, this incompatibility causes a weak fibre/matrix interface, which consecutively leads to diminished mechanical properties of the biocomposites. Therefore, it is important to ensure a good adhesion between matrix and fibres to enhance the mechanical strength of NFRPC.

This study aims to provide an overview of the *greener surface treatments* without use of hazardous chemicals, with emphasize on the enzymatic surface modification of natural fibres. The effectiveness of the treatment on the mechanical properties of the resulting NFRPC is also reviewed.

Environmental impact of NFRPC is another important issue addressed in this review. A comparison of the environmental impacts between the NFRPC and SFRPC applied in the automobile sector, based on LCA studies, will be traced.



1. Introduction

Guidelines for the development of the 21 century generation of materials, products and processes are sustainability, industrial ecology, eco-efficiency and green chemistry.

We assist to an increasing pressure to develop novel bio- and other innovative technologies aiming to rescue extensive dependence on fossil fuel. Synthetic fibres like glass, carbon and aramid are well established reinforcements in polymer composites due to their high stiffness and strength properties [1].

Despite this, serious drawbacks such high processing cost, machine abrasion, high density, health hazards when inhaled and most significant, high energy consumption, non-biodegradability and -recyclability, coupled with severe environmental legislation, shifts attention from synthetic to natural fibres, from Synthetic Reinforced Polymer Composites to Bio- Composites or Natural Fiber Reinforced Polymer Composites (NFRPC) [2]. Besides, not all applications are addressed for structural or load –bearing goals, thus utilizing high- performance fillers for low and non-load bearing are over performance and not cost-effective [3]. Bio-composites defined as composite materials in which at least one of the component is derived from natural resources, can be classed in three subgroups [4]:

1. Petroleum derived polymers reinforced by natural fiber which are non-biodegradable
2. Biopolymers reinforced by natural fibers known as green composites (biodegradable)
3. Biopolymers reinforced synthetic fibers (i.e. glass, carbon etc.) (non-biodegradable).

On the other hand, various industrial application, including automotive, require characteristics that are not met by neat materials. Thus composite materials can be appropriate to fulfill the industrial requirements due to their proper mechanical properties through the synergetic action of the components, matrix and reinforcement, which support the load stress [5].

Under these circumstances, natural fiber composites (NFRPC) as environmentally appealing materials are emerging as a viable alternative to the glass-reinforced or carbon reinforced polymer composites. The final properties and performance of engineering products manufactured from NFRPC depend upon the characteristics of their individual constituents as well as their compatibility and matrix/filler interface.

Currently, NFRPC occupy several industrial area that proofs the potential of natural fibres as replacements for their synthetic counterparts for various application. The fields in which natural fibre composites are being broadly employed include the automotive and construction sectors [6]. Low density and satisfactory high specific properties of natural fibers are features that recommend NFPC for application in the transportation, particularly automotive industry. Attempts for their implication in automobile sector dates back in the 90s, when Mercedes -Benz manufactured door panels containing jute fibers. Flax has been the most relevant natural fibre for the German automotive industry for years, hemp are the second most important natural fibres [7].

Nowadays all the main international automotive manufacturers have adopted in their new models NFRPC for interior and exterior vehicle parts such door panels, package trays, trunk liners, seat backs, boot lining, hat rack, spare tyre lining, headliner panel, boot lining, noise insulation panels, moulded foot well linings, door trim, Interior door paneling, windshield, dashboard, back cushions, parcel shelf, insulation, rear storage shel, door trim panels (PP matrix with NF), engine or transmission cover side and back, spare wheel compartment cover, to name a few [6, 8 - 12].

This shift was prompted by the European Union End of Life Vehicle Directive (2020) imposing that 80wt% of a waste vehicle has to be reused, recovered or recycled [13].

Composite materials find their application also in aircraft, railway and truck industry [14]. Common applications of natural fibre reinforced composites include also household products and furniture, electrical and electronics, sports and leisure items [15] food industry [16] and medical applications [17].

Despite their numerous and manifold uses, there are still obstacles that are obstructing their growth development inside industrial area, particularly for structural purposes.

Apart from synthetic fibres, most natural fibres are relatively hydrophilic, have a rough surface and are physico-chemically heterogeneous. These characteristics exert a strong influence on the fibre-matrix interfacial adhesion in the composite. The poor interfacial bonding leads to defective mechanical properties of the biocomposite [18].

This paper reviews the latest trends in green surface treatments of natural fibers without use of hazardous chemical substances with focus on biotechnology as a viable alternative to the physico-chemical approaches. In this regard, the importance of surface modifications through enzymatic treatment and the resultant enhancement in the properties of the composites are discussed.

An exhaustive approach of the natural fiber structure, chemical composition and properties tries to underline the interdependence existing between these factors. An understanding of the interconnection between the presence of different chemical constituents in the fiber and their influence on the fiber properties and later on upon the NFRPC, represent the key drive for a successfully enzymatic treatment and in turns improved properties of the composite materials for specific purposes. Environmental impact of NFRPC is another important topic addressed in this review.

2. Natural fibers

2.1. Microstructure and properties.

Natural fibres encloses all types of fibres that occur within nature, and are found in vegetables respectively plants (cellulose fibres), animals (protein fibres) and minerals (asbestos). A natural fibre may be further defined as a flexible material which large aspect ratio and high tensile strength.

Even if fibrous materials exist in abundance in nature, particularly cellulosic types like cotton, wood, grains, and straw, not all materials can be spooned into fibres for textile products or other industrial applications. Aside from economic perspectives, the appropriateness of a fibre for commercial purposes is driven by its attributes like length, strength, pliability, elasticity, abrasion resistance, absorbency and surface properties.

Between the plant fibres, lignocellulosic fibres, especially bast fibres (flax, hemp, jute, kenaf, ramie) are by far the most prominent group of natural fibres used as substitute for the synthetic fibres in natural fiber composites in automotive field due to their specific strength, modulus characteristics and availability. Their high content of cellulose make them appropriate for stress-bearing applications [8]. So this review is confined only to plant-based fibres since lignocellulosic fibres accounts for the most technical application.

Lignocellulosic fibers might be considered themselves as composites material due to their cellular structures consisted of cells that contains crystalline and amorphous regions of cellulose linked together through an amorphous matrix of lignin and hemicellulose and pectin. Cellulose is the main constituent of plant fibers, although the percentage composition of pure cellulose, hemicellulose, lignin, pectin and other organic compounds such waxes, fatty acids, fats, pectins varies for different fibres [19].

The hemicellulose molecules are hydrogen bonded to cellulose and act as cementing matrix between the cellulose microfibrils, forming the cellulose – hemicellulose network, which is considered to be the main structural component of the fiber cell. The hydrophobic lignin grid activates as a binder agent of the cellulose/hemicellulose composite, being responsible for the rigidity and low elasticity [20]. In addition, the low elasticity is a result of a low value of microfibrillar angle of the fibers [21].

Furthermore, the cell wall in a fiber is not a homogenous membrane. Actually, each fiber has a complex, stratified structure comprising a thin primary wall surrounding a secondary wall. This secondary wall is an assembly of three layers and the thick middle layer is responsible for the mechanical properties of the fiber [20]. The middle layer has a complex structure composed by a series of helically winding microfibrils along the hollow fibre axis. Microfibrillar angle values differs from fiber to fiber and provide mechanical strength to the fiber. The spiral orientation of the microfibrils to the fiber axis increases the ductility of the fibers [22, 23].

Selected physical and tensile properties of various plant fibers and synthetic fibers are listed in table 1 [8, 24-26]. It can be seen from table 1 that glass fibers have higher tensile strength and higher Young's modulus than cellulose-based fibers. However, comparing the lower density values of the lignocellulosic fibres to the one of glass, it is obvious that the specific tensile strength and specific Young modulus of both types of fibres are comparable. This is an important aspect for automotive industry where weight reduction is a priority [19, 27].

Table 1. Mechanical properties of lignocellulosic fibers

Fibre	Density (g/cm ³)	Diameter (μm)	Tensile strength (MPa)	Stiffness/Young's modulus (GPa)	Specific tensile strength (MPa/g/ cm ³)	Elongation at break (%)	Specific Young's modulus (GPa·cm ³ /g)
Flax	1.38 - 1.52	5 - 600	343 - 1830	27 - 100	227 -1220	1.2 - 3.2	18 - 53
Kenaf	1.2	12 - 36	295 - 930	22 - 60	246 - 993	1.6 - 6.9	18 - 50
Jute	1.23 - 1.5	5 - 200	187 - 800	10 - 55	140 - 610	1.16 - 3.1	7.1 - 39
Hemp	1.35 - 1.51	10 - 500	550 -1110	30 - 70	210 - 740	1.6 - 4.5	20 - 47
Ramie	1.44 - 1.55	18 - 80	400 - 938	44 - 128	258 - 620	1.2 - 4	29 - 85
Coir	1.1 - 1.46	7 - 460	130 - 580	4 - 62	92 - 180	15 - 40	3.3 - 5.2
Sisal	1.2 - 1.5	7 - 200	468 - 855	9 - 28	55 - 610	1.9 - 7	6 - 20
E glass	2.5 - 2.55	15 - 25	200 - 3500	68.9 - 73	666 -1400	2.5 - 3	29

Lignocellulosic fibres proved to be an appropriate reinforcement for structural composites but they do expose some deficiencies emerging from their inherent nature. The tensile properties values comprised in Table 1 reveal a large variability even inside the same type of fibre. This variation can be ascribed to the innate scatter of the fibre properties and test methods applied for tensile properties measurement of single fibre [8, 27]. Strength of vegetal fibres is controlled by a complexity of factors including: the ratio and perfection of crystalline cellulose in the fiber, fiber asymmetry (cellulose molecular chain length-to-width ratio), polydispersity and the cellulose microfibril angle measured to the fibre axis [8, 28].

Further, the length of natural fibre and orientation is an argument for their selection in composites for automotive application [29] since the use of long fibres in composites imparts improved mechanical properties and capability to adhesion over the matrix to the fiber [30].

2.2. Chemical composition of natural fibre.

The chemical constituents of the natural fibres such cellulose, hemicellulose and lignin exert a significant influence upon the overall properties of the fiber and thus upon their applications [19, 31]. As follows, hemicellulose is responsible for the biodegradation, moisture absorption, and thermal degradation of the fiber as it shows reduced level of resistance whereas lignin is thermally stable but responsible for the UV degradation.

Figure 1 exemplifies the interdependency of the natural fibres properties on their major chemical components [32].

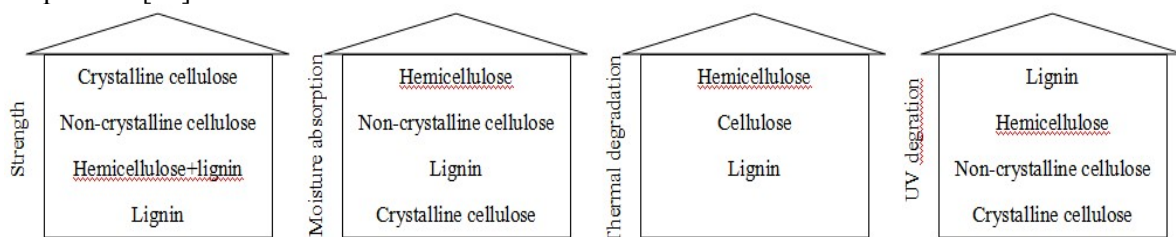


Figure 1. Influence of chemical components upon the properties of natural fibres

The percentage composition of each of these components varies on the type of fiber for different fiber. As a general remark, increased cellulose content will increase tensile strength and Young's modulus of fibers.

3. Natural Fibres Reinforced Polymer Composites – Advantages and challenges

A significant application area of natural fibres is their use as reinforcement for synthetic polymers in the automotive industry. Prior the use of a material in technical application, it must go through an extensive design process, which covers material selection, the generation of a part geometry and calculation of part performance. Since material costs usually exceed 50% of the global production costs, material selection is of utmost relevance, taking into consideration technical and economic requirements. Thus natural fibres will have to compete with materials that are appropriate for a specific technical application. Actually, the criteria for their selection relies in their performance in all aspects which implies a good understanding of their properties, the root cause for their weaknesses and improvement possibilities. The absence of reliable material data for selection and design is the major obstacle for natural-fibres-reinforced materials [33].

A satisfactory prediction of the response of a composite part involves an accurate prediction of the properties of the components (fibre, matrix and fillers), level of adhesion between them and the distribution of the constituents within the composite.

Since synthetic fibres and matrices are usually manufactured under controlled circumstances, their final chemical and physical characteristics are easier to predict. Thus, they can be tailored to have specific properties according to their final application. Unlike synthetic fibres, vegetable fibres, are deriving from nature. Thus, a comprehensive knowledge of their characteristics is more difficult to acquire [5].

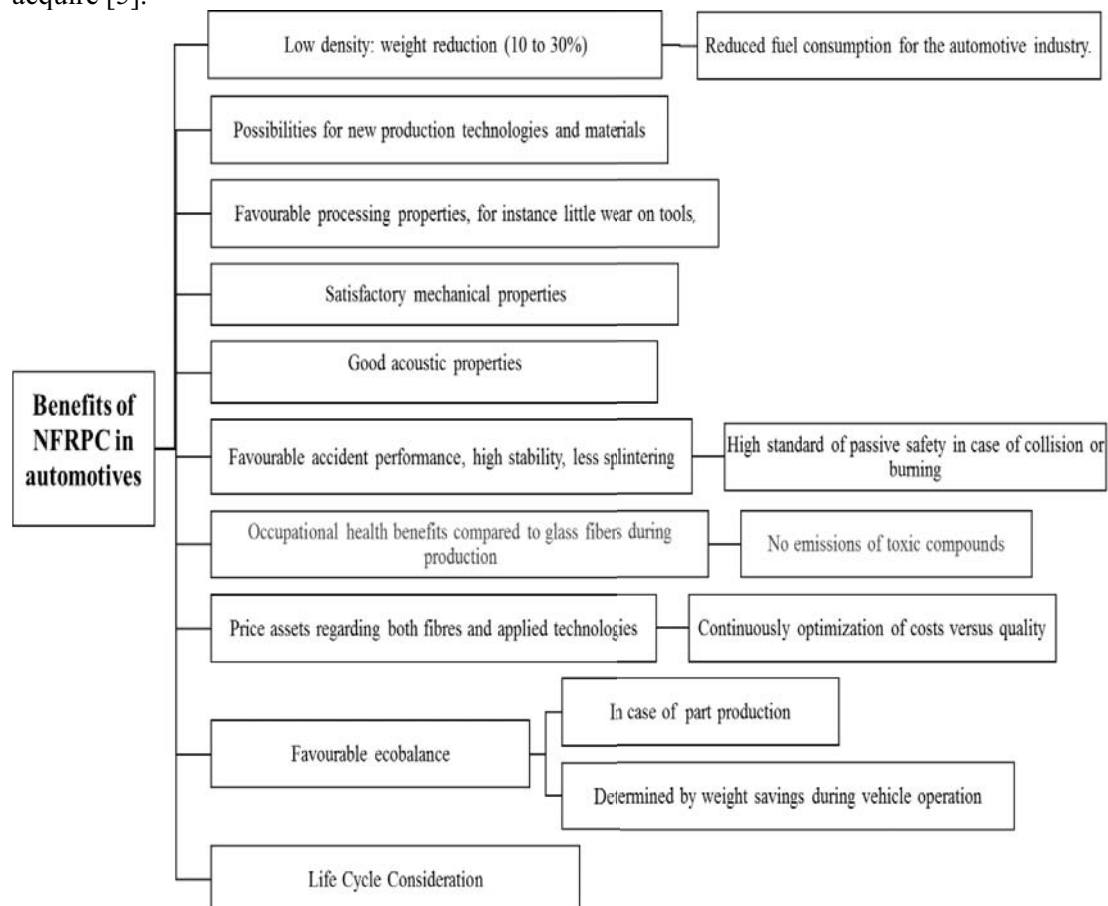


Figure 2. Main benefits of NFRPC in automotive

The prime driving forces for car –makers to replace Synthetic fibre reinforced polymer composites with NFRPC are depicted in fig. 2 [7].

In addition, several factors that are endorsing the application in the automotive sector refers to:

- Reduction of greenhouse gas emissions [34]
- Competitive pricing [11]
- Natural fibers are renewable resources
- Safety, Crashworthiness [35]
- Technical benefits (lower energy loss, better wear protection, expanded lifetime of tools
- Socio- economic benefits (expended usage of NFC in automotive will create new jobs in agriculture [36, 37].

Although these benefits are well known to the industry, natural fiber polymer composite application has been hindered by some drawbacks. Compared to their synthetic counterparts, following aspects have to be taken into consideration [38, 39]

- Structure is highly inhomogeneous
- Fibers discontinuity that causes lower performance of their composites
- Lower durability – possibility to improve considerable
- High moisture absorption which causes swelling
- Lower strength, especially impact strength
- Large scatter of the properties
- Limited thermal stability.

Parameters controlling the mechanical performance of NFRPC are extensively discussed in literature. The main factors in this regard include fibre selection (type, harvest time, extraction, fibre content), matrix selection, fibre dispersion, interfacial strength, composite manufacturing process and porosity [38].

4. Biotechnological procedures for interfacial adhesion improvement

The nature of the fiber-matrix interface exert a critical influence upon the overall properties of composite. The fiber-matrix adhesion and nature of the interphase control the stress transfer efficiency between fiber and matrix, stress distribution, mechanisms of damage propagation and accumulation. Besides the reinforcement of high-strength fibers, high mechanical performance of the composite material is expected in case of a strong fiber-matrix adhesion [40-42].

Natural fibres have some deficiencies due to their strong hydrophilic character which engenders two considerable restrains when used as reinforcement in biocomposites. These include: first, their accentuate sensitivity to water and moisture which provoke in composite a diminishing of mechanical properties upon moisture absorption; second, their incompatibility with the hydrophobic polymeric matrix used, which in turns leads to weak interfacial adhesion [43]. Thus, in several cases NFRPC do not exhibit the same grade of performances compared to the glass –fibers reinforced composites [44].

To overcome these major impediments associated with improper fiber performance, lignocellulosic fibers are subjected to various surface changes. Generally, the adhesion at the composite interface can be ascribed to the following major interactions: physical adhesion related to wettability and compatibility of the fibre - matrix complex, which are controlled by the surface energies of the materials, chemical bonding, and mechanical interlocking build up on the uneven fibre surface [40, 45].

Various modification strategies have been intensively discussed in literature. Processing and surface treatments to compose the interface in NFRPC can envisage either interface engineering trough matrix or fiber modification and the use of interface active additives [46]. Most applied approaches mentioned in literature for improved interface adhesion comprise physical and chemical modification [18, 20]. Chemical approaches are more discussed within the literature than physical with superior improvements obtained till now. Even though positive effects were attained on NFRPC, the applied physical and chemical methods boost the risk for fiber degradation and consecutively to augmentation in the processing cost. Furthermore, it is difficult to solve all the deficiencies associated with NFRPC

using one method [44]. A solution to improve interfacial adhesion in composite and to prevent fiber chain degradation in natural fibre is the use of biological agents, such as fungi and enzymes [47]. Currently, the application of biotechnology in the field of textile and natural fiber modification is well established [48, 49, 50].

The major reason for embracing this technology relies in the high reaction specificity of the enzyme which leads to non-destructive transformation at the fiber surface. Besides, enzymes act under milder condition as compared to chemical methods, decreasing water and energy consumption, producing less toxic wastewaters and are biodegradable [51, 52].

Enzymes used for surface modification of natural fibres belong to the classes of hydrolysis and oxidoreductases. Enzymatic treatments proofed their effectiveness for lignocellulosic fibers in the retting stage [53] as well as in surface modification procedures [54]. Biotechnological retting using specific enzymes is a promising alternative of improving the traditional retting process due to greater control of processing parameters. Thus, the enzymatic retting preserve the natural fiber structure and result in superior mechanical properties compared with chemical retting, which damages structure and lessens quality [55].

This review is conceived with the biotechnological procedures for fiber surface modification of bast fibers, for improved interfacial strength. Some representative literature references are discussed in detail. There are several reports describing improvement of composite properties ascribed to enzymatic treatment of natural fibres. Z Saleem *et al.* [56] report the effect of pectinase treatment of hemp on mechanical properties of fibre reinforced polypropylene. Their results support evidence for an effective refinement of hemp fibres when treated with a commercial pectinase. Although the structural changes caused by the pectinase action altered the mechanical properties, the composite prepared using enzymatic treated hemp present improved tensile and flexural characteristics. Actually, the fibre refinement results in separation of technical fibres into smaller bundles and single fibre cells which in turns means an increasing in surface to mass ratio of the fibres and an enlarged interfacial area between fibre and matrix. Further studies reported that the enzymatic treatment of natural fibres leads to increased surface area and increased interfacial bonding coupled with increased impact properties [57].

Karaduman *et al* [58] also confirmed these findings by tracing a comparison between alkaline and enzyme-treated jute fibres on the mechanical properties of jute fiber-reinforced polyester composites. A set of various enzymes (pectinase, laccase, cellulase and xylanase), enzyme combinations and treatment time according to experimental design were employed in the study. Tensile and flexural properties measurement were carried out to evaluate the efficacy of the treatments. As a result of the enzymes action, pectin, hemicelluloses and lignin were removed from the fiber bundle interface. This action reduced the technical fiber diameter and hence increased the fiber aspect ratio which enables better fiber-matrix adhesion and improved mechanical properties of the composites [58].

Dong A *et al* [59] investigated the possibility to obtain an hydrophobic jute fabrics via the laccase-mediated grafting of octadecylamine (OA) on the lignin moieties. The hydrophobized jute was used as the reinforced material of the polypropylene (PP) matrix composites to improve the interfacial compatibility between the fiber reinforcements and the hydrophobic synthetic polymer. To evaluate the effectiveness of the enzymatic treatment on jute fabric/ PP composites, the tensile properties, dynamic mechanical performances and fracture surface of were analyzed [59]. Through laccase-mediated grafting of OA, modified jute gained excellent water repellency. As a consequence of the specificity of laccase to substrates and the surface modification of the enzymatic grafting no evident decrease of thermal properties was noticed. In addition, the tensile strength, tensile modulus as well as the breaking elongation of the hydrophobized jute/PP composites were enhanced. The treatment lead also to an improved interfacial adhesion between the jute reinforcement and the PP resin [59]. Laccases – assisted grafting process jute fibers using different chemical compounds proofed its efficiency, too [60, 61].

After the enzymatic graft modification the jute/PP composites present improved tensile and dynamic mechanical properties and good water absorption repellency. Besides, the hydrophobic modified jute represent a prime condition for good interfacial compatibility.

Improved surface properties of the natural fiber reinforcement can be achieved also through removal of amorphous constituents in order to gain as much cellulose as possible with higher degree of crystallinity. For this purpose the effect of various enzymatic systems on the surface chemical, morphological and thermal properties of bast fibres was studied [62]. Flax and hemp fibres were subjected to treatment with hemicellulases, pectinases and oxidoreductase. Removal of amorphous hemicellulose from the fibre surface and following exposure of the crystalline cellulose caused a lower contact angle for all the treated samples.

Other beneficial effects of enzymatic treatments are associated with a homogeneous cellulose surface for subsequent activation [63] and an improved thermal stability due to the removal of the thermally labile pectic and hemicellulosic fiber constituents. Compared to hemp fibres, flax is more susceptible to enzymatic degradation. The results confirm the enzymatic treatment as an inexpensive and environmentally appealing alternative to improve the surfaces of natural fibres for composite applications [62].

5. Environmental performance of NFRPC

In order to reduce environmental impact the automotive industry has embraced several directions: development of sophisticated emissions control technology that provide much cleaner vehicle with catalytic converters reducing smog-forming emissions for car; development of alternative fuels, hybrid technology [64]. Accordingly, considerable efforts are also directed towards improving the fuel efficiency of vehicles as required by global regulations. These target can be fulfill by weigh reduction because lower weight vehicles combine the use of less fuel with fewer carbon dioxide emissions [34]. Thus, NFRPC application in automotive has increased significantly mainly due to their ability to meet diverse requirements like significant *weight savings*, specific strength, stiffness and *environmental benefits*.

Natural fiber composites are claimed to offer multiple environmental advantages such as: reduced dependence on non-renewable energy/material sources, lower pollutant emissions, lower greenhouse gas emissions [65] enhanced energy recovery and end of life biodegradability of components [39].

However, all these assertions should be supported by quantitative analysis. In this regard, Joshia *et al.* analyzed three comparative Life Cycle assessment (LCA) studies to assess the overall environmental performances. Results reveal that in their specific applications, analyzed in the study, NFRPC composites are environmentally superior to glass fiber composites on most midpoint indicators [66].

Drivers of superior environmental performance depicted from the comparative analyses are as follows [66]:

- Less environmental impacts of natural fiber production compared to glass fiber production
- Substitution of matrix (synthetic polymer) by higher content of natural fiber in the composite
- Improvement of fuel efficiency and lower use phase emissions due to weight reduction
- Energy and carbon credits from end of life incineration of natural fibers [39].

Yet, these benefits are counter-balanced by the environmental burdens caused by the use of fertilizer in natural fibre cultivation. Thus, the high nitrate and phosphate emissions can lead to increased eutrophication in local water bodies. A reduced operating lifetime compared to the glass fibre composites affect the environmental superiority of natural fibre composites [66].

In their study, George M *et al.* highlight also the environmentally superiority of NFRPC compared to their synthetic counterparts. For this purpose, hemp fibres modified by chemical treatments for composite application were evaluated for their environmental impact. Two novel acid sulfonic procedures and conventional chemical methods were applied for surface fiber improvement. Previously, a comparison between the novel and established chemical procedures in terms of environmental burdens was traced. The new methods had a comparably lower environmental impact than the conventional ones. Furthermore, all the chemical methods proved to have environmental

lower footprint than the glass fiber reinforced polymer. Even with a chemical treatment the NFRPC were superior to glass fiber. These assumptions are based on LCA outcomes [67].

6. Conclusion

Fibres as reinforcement present some features that will interfere in the final properties of composites. Vegetal fibers, due to hydrophilic characteristic, are incompatible with most hydrophobic matrixes, weakening the interfacing of composite.

To overcome the hydrophilic/hydrophobic incompatibility, the enzymatic surface treatments envisage either the removal of amorphous constituents aiming to get as much cellulose as possible with higher degree of crystallinity or the mediation of a grafting process. The results obtained in several studies proved the potential of enzymatic surface treatment of natural fibres as an effective and environmentally friendly tool for natural composites with improved interfacial adhesion and mechanical properties.

Additional, studies assessing life cycle environmental performance of natural fiber composites and glass fiber reinforced composites considered for specific applications, reveal the environmental superiority of natural fiber composites.

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