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# Natural Fibres Reinforced Green Composites

## *Z naravnimi vlakni ojačeni zeleni kompoziti*

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### Abstract

Composites made from natural fibres and various bio polymers have recently attracted great attention. In this paper, the reasons for the popularity of green composites are highlighted. The disadvantages and modification methods of natural fibres are then summarized. Additionally, biodegradable polymers and a brief survey of the research work are pointed out.

Keywords: natural fibres, composite materials, biodegradable polymers, ecology

### *Izvleček*

*Kompoziti iz naravnih vlaken in različnih biopolimerov v zadnjem času zbuja veliko pozornost. V članku podani pregled osvetljuje vzroke za priljubljenost zelenih kompozitov. Povzeti so slabosti naravnih vlaken, metode njihove modifikacije in pregled biorazgradljivih polimerov ter kratek pregled izvedenih raziskav na tem področju.*

*Ključne besede: naravna vlakna, kompozitni materiali, biorazgradljivi polimeri, ekologija*

## 1 Introduction

A green or an eco/ecological composite may contain natural fibres and natural polymers, or it can be a combination of natural fibres and a biodegradable polymer matrix [1]. Actually, natural raw materials have been used in composites since 1850. By combining shellac-resin and sawdust little boxes for photographs were produced until 1870. According to Müssig, in the 1920s and 1930s, the first natural fibre composites were manufactured in aircraft construction with the aim of creating lighter parts for primary structures. In the late 1930s, Henry Ford promoted their application in the automotive sector [2]. Automotive and aerospace industries still demonstrate an interest in using more natural fibre reinforced composites [3].

The reasons for this great interest are of course the environmental advantages and biological degradability of natural fibres. For example, if flax is burnt after having been used, no residues remain [4]. Natural materials like wood chips and flour are also

widely used because of their eco-friendly characteristics as fillers within thermoplastics in automotive and building applications [5]. Even though wooden materials are cheaper than natural fibres, it is expected that in the near future, due to extensive native forest destruction, their price will increase to a point at which they will be no longer able to compete with natural fibre composites. This is already the case for some native forest-depleted countries [6].

Other advantages of natural fibres in composite reinforcement are:

- natural fibres are abundant and renewable [7];
- production of natural fibres results in lower environmental impacts compared to glass fibre production [8];
- lower weight of natural fibre composites improves fuel efficiency and reduces emissions during the use phase of the component [9]. The weight reduction when conventional composite materials are replaced with natural fibre composites can reach even 26%. More flax fibres have been progressively incorporated into natural fibre

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automotive components that require 83% less energy and are 40% less expensive than glass fibre components [10];

- end of life incineration of natural fibres results in energy and carbon credits;
- reduced tool wear [11];
- they are safer especially in automobile interiors as the fractures of natural fibre composites are not as sharp as the fractures of glass fibre composites [12].

Natural fibres used in composite reinforcement are split into four categories; seed, bast, leaf and fruit. Cotton is a seed fibre. The bast fibre group includes kenaf, hemp and flax, while sisal may be considered a leaf fibre. On the other hand, coir and banana are vegetable fibres [13]. According to Zampaloni 2007, bast fibres exhibit superior flexural strength and modulus of elasticity (MOE), whereas leaf fibres show superior impact properties [3]. Tensile strength and elastic modulus values of these fibres are given in Table 1.

*Table 1: Tensile strength and modulus of elasticity of natural fibres used in composite reinforcement*

Fibre Type	Tensile Strength (MPa)	Modulus of Elasticity (GPa)
Flax [10]	1100	100
Cotton [10]	287-597	5.5-12.6
Jute [6]	550-900	17-26
Sisal [6]	400-700	9-20
Ramie [6]	870	128
Coir [6]	106-270	3-6
Banana [6]	529-800	7.7-32

Beyond the advantages mentioned below, natural fibres have some limitations and drawbacks, when they are used as reinforcements in composite materials, such as:

- the fibres are hydrophilic;
- the compatibility between hydrophilic fibres and hydrophobic matrices is low;
- the fibres are not resistant to high temperatures (>200 °C);
- the fibres are short;
- the quality and consistency of properties is affected by factors which are hard to control such as climate impacts during growth and harvesting [14].

## 2 Modification of natural fibres in composite materials

The characteristics of composites depend on:

- the properties of fibre,
- the properties of matrix material,
- interfacial compatibility of fibre and matrix.

Natural fibres contain hydroxyl and other polar groups in their constituents [7]. This feature becomes a disadvantage when these fibres are used in composite materials since they meet with non-polar polymers. Polar groups are hydrophilic which causes the absorption of water in natural fibres. This nature leads to incompatibility and poor wettability in a hydrophobic polymer matrix and weak bonding in the fibre/matrix interface [11].

The surface adhesion between the fibre and the polymer plays an important role in the transmission of stress from the matrix to the fibre. If bonding between the fibre and the polymer matrix material is weak, then failure is inevitable in ultimate composite. The absorption of water in the pores and amorphous regions of natural fibres serves to reduce interfibrillar cohesion and to relieve internal fibre stresses [15]. Furthermore, the high moisture absorption property of natural fibres makes them less attractive for exterior applications [7], decreases their dimensional stability [6] and tends to rotting [16].

There are several methods existing which intend to improve surface adhesion between natural fibre and polymer matrix material. The basic objective, however, remains the same: to remove surface contamination and to provide an intimate contact between the surfaces. The simplest way is to roughen the surface so as to enhance the contact area and facilitate mechanical interlocking [11]. Modification methods are classified as physical and chemical.

### 2.1 Physical methods of modification

Physical treatments change structural and surface properties of natural fibres and thereby influence their mechanical bonding with the matrix [6]. Physical methods involve surface fibrillation, plasma, corona, dielectric barrier techniques etc.

Plasma can be defined as a partially ionized quasineutral gas. In this ionized gas there is the balance between the densities of negative and positive particles in macroscopic volumes and time. Plasma components include ions, electrons, UV and vacuum UV radiation. Plasma is thought to bring physical

modification on the surface through roughening the fibre by the sputtering effect, producing thus an enlargement of the contact area that increases the friction between the fibre and the polymer.

Corona and dielectric barrier techniques are non-thermal plasma treatments. Corona discharges are relatively low power electrical discharges that take place at or near atmospheric pressure. The corona is generated by strong electric fields associated with small diameter wires, needles, or sharp edges on an electrode. Corona treatment is a difficult technique to be used on 3D objects. This limitation has highlighted the need for alternative, more 3D 'compliant' surface modification techniques such as the ozone and/or ultraviolet radiation (e.g. UV) light [11].

### 2.2 Chemical methods of modification

The compatibility and dispersability of fibre and matrix can be improved by developing a hydrophobic coating of a compatible polymer on the surface of a filler before being mixed with polymer matrix. Generally, coupling agents facilitate the optimum stress transfer at the interface between the fibre and the matrix. Coupling agents are molecules having two functions. The first is to react with OH groups of cellulose and the second is to react with functional groups of the matrix [7].

There are many types of coupling agents including surface-active agents and reactive chemistries, at times referred to as functional modifiers. Surface-active agents are materials that increase interfacial adhesion by acting as a solid surfactant and that do not form covalent bonds to the polymer matrix. Materials that form covalent bonds to either the fibre or the polymer matrix can be reacted in-situ during processing [17].

The selection of a coupling agent that can combine both strength and toughness to a considerable degree is important for a composite material. The most common coupling agents are silane, isocyanate and titanate based compounds, the chemical composition of which allows them to react with the fibre surface, which forms a bridge of chemical bonds between the fibre and the matrix [7].

Those options (physical and chemical modifications) are not attractive, even in terms of cost-benefit ratio, when the composites are designed for low strength-low cost applications such as transit panels, because of the significant cost increase that might result from the use of chemicals or treatment apparatus [6].

## 3 Matrix materials

Matrix materials are divided into two categories as thermosets and thermoplastics. Thermosets cannot be melted once they have been cured since they are chemically crosslinked. But these matrices generally have quite good mechanical properties. Most common thermoplastics have low transition temperature, giving them quite low stiffness at room temperature and above. The low T<sub>g</sub> also makes them very brittle at low temperatures, however, the lack of cross-linking is one of the big advantages of thermoplastics which makes them reshapeable and re-meltable [14]. Thermoplastics also offer many advantages. One of the advantages of thermoplastic matrix composites is their low processing costs. Other advantages are the design flexibility and the ease of moulding complex parts. Simple methods such as extrusion and injection moulding are used for the processing of these composites [18].

Most commonly used thermoset polymers in natural fibre reinforced composites are polyester, epoxies and vinylester. While natural fibres have been traditionally used to fill and reinforce thermosets, natural fibre reinforced thermoplastics, especially polypropylene composites, have attracted greater attention due to their added advantage of recyclability [8]. Other thermoplastics like polyethylene (PE) and polystyrene (PSI) have also been used in ecological composites.

No matter which polymer is used, high temperatures must be avoided due to the possibility of fibre degradation. When working with natural fibres, the processing temperature must be below 200°C [18]. This also restricts natural fibre composites to relatively low temperature applications [19].

Low twisted yarns display a very low strength when tested dry in the air and, therefore, they cannot be used in processes such as pultrusion or textile manufacturing routes. On the other hand, by increasing the level of twist, a degradation of the mechanical properties is observed in impregnated yarns (e.g. unidirectional composites) similar to off-axis composites. Therefore, an optimum twist should be used to balance processability and mechanical properties [20]. On the other hand, strands of natural fibres are usually heavily twisted, and that can hinder infiltration or impregnation of the resin matrix into the fibre strands [21].

### 3.1 Biodegradable polymers

There are many different polymers of renewable materials. Biodegradable plastics can be based on natural or synthetic resins. Natural biodegradable plastics are based primarily on renewable resources (such as starch) and can be either naturally produced or synthesized from renewable resources [22].

PLA is a good example of thermoplastic biopolymer that is used today. Its basic monomer is lactic acid, which is derived from starch by fermentation. PLA is and was frequently used for biodegradable packing materials. However, numerous tests have shown that PLA is also suitable as a matrix for embedding fibres in composites [23]. Other studied biopolymers have been soy-oil based epoxy, starch, polycaprolactone (PCL), polyhydroxybutyrate (PHB), modified cellulose, acetic acid, polyester amide, natural rubber and gluten [22, 24].

Polyesters play a predominant role as biodegradable plastics due to their potentially hydrolysable ester bonds. Biodegradable polyesters which have been developed commercially and are in commercial development are PHA – polyhydroxyalkanoates, PHH – polyhydroxyhexanoate, PHB – polyhydroxybutyrate, PHV – polyhydroxyvalerate, PLA – polylactic acid, PCL – polycaprolactone, PBS – polybutylene succinate, PBSA – polybutylene succinate adipate, AAC – aliphatic–aromatic copolyesters, PET – polyethylene terephthalate, PBAT – polybutylene adipate/terephthalate and PTMAT – polymethylene adipate/terephthalate [22].

The problems with most of these polymers have been poor commercial availability, poor processability, low toughness, high price and low moisture stability [25].

## 4 Preforms

Natural fibres such as flax are usually available as short fibres. These short fibres are suitable for injection moulding process and for producing non woven mats. In both processes, short fibres are randomly organised which causes relatively poor mechanical properties in resulting composites [20].

There are several studies available concerning the mechanical properties of green composites. A brief survey of the research work published in the field of natural fibre reinforced composites is given in Table 2. According to the literature, preform types have been generally used in the form of nonwoven/fibre mat.

## 5 Conclusion

The aim of this article is to give a short overview of the reasons for the usage of green composites, disadvantages of natural fibres and their modification methods, matrix materials and research activities associated with these composite materials.

The research showed that there are many types of natural fibres and biopolymers available for green composites all of which have different characteristics and benefits. Although modifications and biopolymers increase the cost of ultimate composite, their environmental returns are undeniable.

Table 2: Literature survey of green composites

Reference	Fibre type	Preform type	Matrix material type	Tensile strength (MPa)	Impact strength (kJ/m <sup>2</sup> )
[7]	100 % flax	nonwoven mat	epoxy	57.79	13.69
[26]	100 % flax	nonwoven mat	modified soy oil	40	
[6]	100 % jute	fibre mat	PES	43.0	
	100 % sisal	fibre mat	PES	34.6	
[27]	100 % flax	fibre mat	Palatal E240-02	47.9	
		fibre mat	Norpol 200-510	71.6	
		fibre mat	epoxy	88.3	
		fibre mat	Norpol 420-100	79.2	
		fibre mat	vinylester	91.2	

[28]	100 % jute	nonwoven mat	PLA	72.7	14.3
[29]	100 % kenaf	fibre mat	PP	28	14
	100 % coir	fibre mat	PP	10	24
	100 % sisal	fibre mat	PP	35	28
	100 % hemp	fibre mat	PP	52	26
	100 % jute	fibre mat	PP	26	17
[30]	100 % flax	nonwoven mat	cellulose acetate propionate		10
[31]	100 % jute	plain weave	PES	23.1	
		flat knit	PES	17.8	
[32]	flax 50 %, cotton 50 %	denim fabric	HDPE	35.8	
[20]	flax	plain weave	vinylester	129	
[33]	100 % flax	fibre mat	PPMA	75	
			PLLA	100	
			PLA	100	
			PHB	40	
			PBS	50	
			PBAT	30	
[21]	jute 45 %, cotton 55 %	plain weave	PES	62.5	
	jute 39 %, cotton 61 %	plain weave	PES	72.1	
	jute 43 %, cotton 57 %	plain weave	PES	73.1	
	100 % jute	plain knit	PES	19.8	
	100 % jute	plain weave	PES	40	
[34]	sisal and silk	hand lay up	PES	18.95	
		hand lay up	PES	23.61	
[3]	kenaf	hand lay up	PP	45	
	kenaf	hand lay up	PP	43	
	kenaf	hand lay up	PP	51	
	hemp	hand lay up	PP	52	
	coir	hand lay up	PP	10	
[2]	cotton ramie	multilayer web	epoxy	28	82
		multilayer web	epoxy	100	38
[35]	sisal sisal	plain weave	PES	28	
		twill weave	PES	33	
[36]	abaca man-made cellulose		PLA	74	5.3
				92	7.9
[1]	cotton	spacer fabric with weft inlay	PLA	30.31	11.18
[37]	flax	spacer fabric with weft in-lay	epoxy	41.45	53.80
	flax	spacer fabric with weft in-lay	vinylester	54.70	49.23
	cotton	spacer fabric with weft in-lay	epoxy	49	19.26
	cotton	spacer fabric with weft in-lay	vinylester	42.41	17.27

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