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UTILIZATION OF AGRICULTURAL WASTE AND WOOD INDUSTRY RESIDUES IN THE PRODUCTION OF NATURAL FIBER - REINFORCED COMPOSITE MATERIALS

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ABSTRACT

Composite materials, based on renewable and biodegradable natural fibers, derived from agricultural waste and wood industry residues, are increasingly utilized in a wide variety of applications. These products represent an ecological and inexpensive alternative to the traditional petroleum-based materials, as they significantly decrease the use of fossil fuels and reduce the greenhouse gas emissions. In addition, these materials have good mechanical properties and require lower consumption of energy for their production. On the other hand, wood-based industries and agriculture produce significant amounts of organic waste and residues which are still underutilized, as low value energy resources and organic waste is commonly disposed of by some of the traditional waste management techniques, such as landfilling, anaerobic digestion or composting. The use of organic agricultural and wood industry waste and residues in production of natural fiber-reinforced polymer composites (NFPCs) is an environmentally friendly, sustainable and economical alternative. This paper represents a review of the possibilities for application of organic waste and residues as reinforcements or additives in NFPCs on the basis of the existing scientific information in the respective field.

Key words: natural fiber-reinforced composites, sustainability, circular economy, wood industry, biocomposites

1. INTRODUCTION

The increased environmental consciousness and shortage of natural resources, as well as the recent strict environmental regulations and unsustainable consumption of fossil fuels, have forced many manufacturing industries to search for new eco-friendly materials from renewable resources, in order to substitute the conventional materials in a number of end uses. This growing need for 'green' materials has led to the increased utilization of natural fibers in production of composite materials. Due to their annual renewability and biodegradability, natural fibers derived from agricultural residues and wood industry waste represent an ecological and inexpensive alternative to the traditional petroleum-based materials, as they significantly decrease the use of fossil fuels and reduce the greenhouse gas emissions. In addition, these materials are easy to process, recyclable, have good mechanical properties and require lower consumption of energy for their production. The use of organic agricultural and wood industry waste and residues in the production of natural fiber-reinforced polymer composites (NFPCs) is a viable, sustainable and economical alternative for replacing harmful synthetic materials in many industrial sectors such as construction, automotive, furniture, etc.

According to the Waste Framework Directive (Directive 2008/98/EC), *waste* means 'any substance or object which the holder discards or intends or is required to discard'. In addition, the

Directive gives the following definition of the term *by-product*: 'substance or object, resulting from a production process, the primary aim of which is not the production of that item'.
The transition to circular economy and the recent targets set by the European legislation have posed greater challenges in relation to waste management. The European Union's approach to waste management is based on the "waste hierarchy" which sets the following priority order: prevention, (preparing for) reuse, recycling, recovery and, as the least preferred option, disposal (which includes landfilling and incineration without energy recovery).
According to the latest statistical data, 16 tonnes of material are currently used per person each year in Europe, of which 6 tonnes become waste. Although the management of that waste has significantly improved in the EU countries, the European economy currently still loses a considerable amount of potential 'secondary raw materials' such as metals, wood, glass, paper, etc. In 2010, total waste production in the EU amounted to 2,5 billion tonnes. From this total only a limited share (36%) was recycled, while the rest was landfilled or incinerated, of which some 600 million tonnes could be recycled or reused. According to some authors (Mantau, 2012) 26 million tonnes of post-consumer wood (wood products that are disposed at the end of their life cycle, e.g. wooden furniture, packaging, doors, windows, various construction materials, etc.) was generated in Europe in 2010. Of these quantities, 7.8 million tonnes were recycled into other products and 10.3 million tonnes were burned for energy in power plants or households. Other about 8 million tonnes were permanently disposed of in landfills or incinerated (not for energy). Large quantities of wood wastes are also produced in the course of primary and secondary wood processing, including bark, shavings, slabs, off-cuts, rejects, wood chips and saw dust. These types of waste, sometimes called wood processing residues, are produced at industrial facilities and are easily collectable and reusable.
Taking into account these figures, it is important to create different applications for the previous waste and residue materials while considering environmental and economic factors.

2. CHARACTERISTICS AND APPLICATIONS OF NATURAL FIBER - REINFORCED COMPOSITES

In the recent years, there have been significant improvements in the properties of the bio-based composite materials and they have been widely used in numerous engineering fields as alternative of the conventional materials. Natural fiber polymer composites are innovative composite material consisting of a polymer matrix reinforced with high-strength natural fibers. The polymer materials in the composition of NFPCs can be divided into two main categories – thermosets and thermoplastics. Thermoplastics include polymers such as polyethylene, polypropylene, polystyrene and polyvinyl chloride, while polyurethane, epoxy and polyesters are examples of thermosets used for production of NFPCs. Thermoplastics have multiple advantages over thermosets such as recyclability and remoldability, and for that they are predominantly used (Clemons, 2008). The main limiting factor of the polymer material in the composition of the NFPCs is the processing temperature which has to be below 200° C to avoid thermal degradation of the natural fibers (Lilholt and Lawther, 2000).

According to some authors (Corbiere-Nicollier et al., 2001) the use of biomass in bio-based composite materials has a much greater potential than the use of biomass for direct heat production or production of biofuels for transport. Studying the life cycle of renewable natural fibers, the same authors revealed that the use of natural fibers as a substitute of glass fibers is beneficial from environmental perspective. Moreover, in some cases the NFPCs demonstrated better performance than the glass-reinforced composite materials (Joshi et al., 2004) and are of much lower risk to human health (Huda et al., 2008). Natural fiber-reinforced composites can be viable alternatives to synthetic fiber-reinforced composites as structural or semi-structural components, especially in lightweight applications (Sathishkumar et al., 2014; Sanjay et al., 2017; Yusriah et al., 2014). Some of the already explored applications of natural fiber-reinforced composites include window and door frames, furniture, railroad sleepers, automotive panels and upholstery, packaging and other applications that do not require very high mechanical resistance, but, instead, significantly reduce the purchasing and maintenance costs (Faris et al., 2014; La Mantia and Morreale, 2011). Natural fiber composite materials in the form of panels and sandwich plates have been used to replace wooden fittings, furniture and noise insulating panels (Mei-po et al., 2011).

The production of NFPCs has been extensively studied in the recent years. Some of the authors studied the so-called wood-plastic composite materials, while others focused their research on jute, hemp, bamboo, sisal and kenaf fiber-reinforced composites. Data about the annual production of natural fibers in presented in Table 1.

Table 1. *Annual Production of Natural Fibers*

Natural fiber	**Annual production, metric tones**
Wood (hardwoods and softwoods)	1 750 000 000
Softwood kraft pulp	26 000 000
Sisal	378 000
Kenaf	970 000
Hemp	214 000
Bamboo	30 000 000
Wheat	720 000 000
Jute	2 300 000

The chemical composition of natural fibers varies greatly between the different fiber types, but the main chemical constituents are cellulose, hemicellulose and lignin. Other constituents include extractives, ash, pectin and waxes (Rowell et al., 2000, Lilholt and Lawther, 2000, Saarimaa et al., 2007, Liu et al., 2012). For example, the chemical composition of hardwood natural fibers is the following: 40-50% cellulose, 23-39% hemicellulose, 20-30% lignin, 2-4% extractives, 0,2-0,4%ash, 0-1% pectins and 0,4-0,5%waxes. Since cellulose is the principal chemical component of plant fibers, with varying amount of hemicellulose and lignin, they are often referred to as lignocellulosic or cellulosic.

The worldwide availability of renewable resources and biodegradability of lignocellulosic fibers, their low cost compared to synthetic fibers, and good specific mechanical properties, have resulted in increased interest towards the industrial utilization of natural fibers in the production of composite materials. In addition, the use of agricultural and wood industry resources has created additional business development opportunities in countries with insufficient fossil fuel stocks, which in turn has provided conditions for sustainable development.

Despite the numerous advantages of natural fibers, there are some limitations that reduce their potential as raw material by compromising the strength of the resulting composite material (Saheb and Jog, 1999; Faruk et al., 2012; George et al., 2001). Some of these disadvantages include incompatibility with the matrix and insufficient adhesion, low water and thermal resistance, susceptibility to microbial attacks, etc. These disadvantages impose some limits on the application of natural fibers in composite materials, but also provide opportunities for researchers to either eliminate or minimize their effects. The main advantages and disadvantages of natural lignocellulosic fibers, related to their utilization for production of composite materials, are listed in Table 2.

Table 2. *Advantages and disadvantages associated with natural fibers*

Advantages	**Disadvantages**
Renewable resources	Inhomogeneous structure of fibers
Lower production costs	Dimensional instability as a negative consequence of water absorption
Good specific mechanical properties	Lower water and thermal resistance
Lower density of composites	Susceptibility to microbial attacks and rotting
Reduced energy consumption during manufacturing	Insufficient adhesion and incompatibility with the polymer matrix
Biodegradability and eco-friendly materials	Degradation and aging
Lower risk to human health	Restricted processing temperature (to avoid thermal degradation)

3. UTILIZATION OF ORGANIC WASTE AND RESIDUES IN NFPCs

The principles of circular economy have imposed new, stricter requirements to all aspects of product design and development. The new legislative requirements in the field of waste management have enhanced the transition to bio economy and have increased the demands for new 'green' materials. In this respect, the recycling and secondary utilization of waste are important issues of economic profitability and sustainable development of woodworking and furniture industries.

Utilization of agricultural and wood wastes and residues in the composition of the NFPCs can be achieved in three main approaches: they can be used as additives (to improve the compatibility between the main components) or as reinforcements of the plastic matrix. Organic waste and residues can also be used as an inexpensive and renewable filler material. The use of organic waste and residues in NFPCs increases the share of raw materials from renewable resources, which results in reduced overall cost of the composite materials. The decreased raw material costs and the significantly improved waste management techniques are one of the most distinctive advantages of organic waste utilization for production of NFPCs.

Different manufacturing methods have been established to produce composites, such as film stacking, vacuum infusion, compression moulding, filament winding, manual winding, resin transfer moulding, injection moulding, etc. (Khondker et al., 2006; Liu and Hughes, 2008; Yan et al., 2012). While selecting a particular manufacturing method, various factors need to be considered, including raw material availability and properties, size and shape of the composite material, economic characteristics of the manufacturing process, etc. (Danni et al., 2014; Mei-po et al., 2011).
Some possible ways for utilization of organic residues in NFPCs are presented in Figure 1.

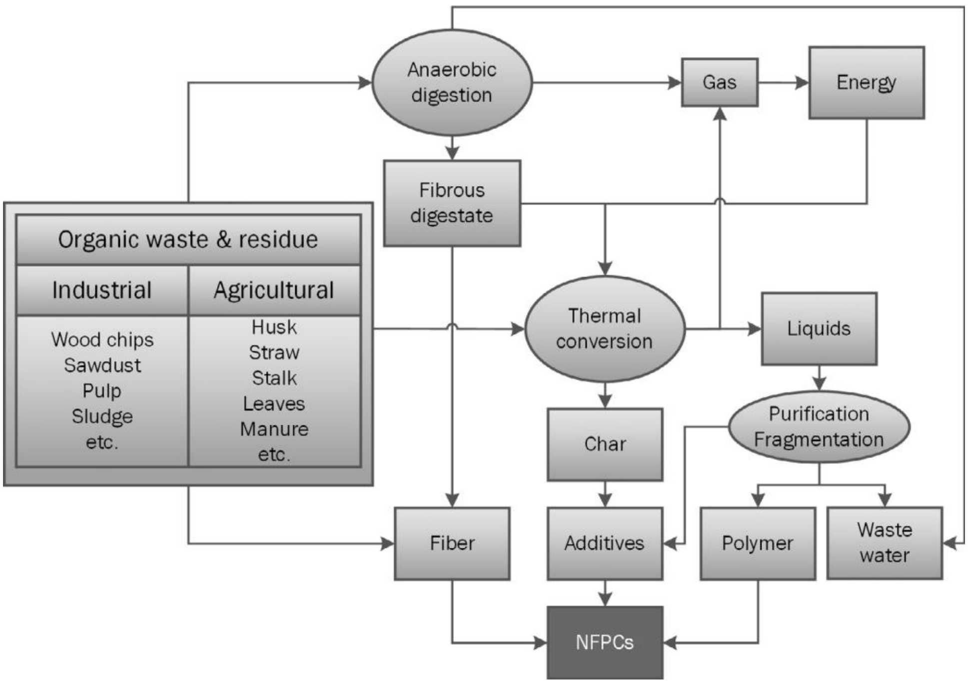

Figure 1. *Possible ways for the utilization of organic waste and residues in NFPCs (Väisänen, T., et al., 2016)*

4. PROPERTIES OF NATURAL FIBER – REINFORCED COMPOSITE MATERIALS

The properties of the natural fiber-reinforced composite materials depend to a great extent on the type of natural fibers (physical and chemical properties), fiber treatment method, fiber orientation and defects, type of polymer matrix, and interaction with the fibers, manufacturing method, additives, etc. (Ku, H., et al. 2011; Hänninen, T., et al, 2012; Clemons and Caulfield, 2006; Shalwan and Yousif, 2013). The bonding strength between fibers and polymer matrix in the composite material is considered to be of great importance for achieving sufficient mechanical properties of the final products. According to many authors (Mutje et al., 2007; Mohanty et al., 2006; Li et al., 2014; Premalal et al., 2002; Joseph et al., 1999), the presence of natural fibers in the polymer matrix normally increases the strength and modulus of the composite materials. Tensile strength is more dependent on the properties of the polymer matrix, whereas the elastic modulus of the composite is primarily dependent on the properties of the fiber (Saheb and Jog, 1999). The ratio of the fiber length to fiber diameter determines the fracture properties of the NFPCs (Stark and Rowlands, 2003). The mechanical performance of the composites depends on the so called critical fiber length (Stark and Rowlands, 2003; Bourmaud and Baley, 2007) which in turn depends on fiber characteristics and shear strength of the matrix-fiber bond. If the length of the fiber is less than its critical length, the matrix-fiber-interface is likely to fail due to the deboning at lower stresses (Stark and Rowlands, 2003; Bourmaud and Baley, 2007).

Some properties of NFPCs, depending on the type of fiber, are presented below. Epoxy resin in ratio 10:1 to fibers is used as matrix. Polymers with cotton fibers at density of 1500 kg.m^{-3} have bending strength of 400 N.mm^{-2} and modulus of elasticity from 5500 to 12 600 N.mm^{-2} (Wambua et al., 2003; Ahmad et al., 2006). Those with the same density but with flax fibers have much higher bending strength up to 1500 N.mm^{-2} and modulus of elasticity of 27 600 N.mm^{-2} (Nabi Saheb and J.P. Jog, 1999). The best result for the modulus of elasticity is obtained when softwood craft pulp is used. Those polymers with density of 1500 kg.m^{-3} have bending strength of 1000 N.mm^{-2} and modulus of elasticity of 40 000 N.mm^{-2} (Hajnalka et. al., 2008). Therefore, in comparison with composites with cotton fibers, the ones with craft pulp have nearly four times better modulus of elasticity.

If a comparison is made between polymers of hemp and kenaf fibers at density of 1460 kg.m^{-3}, the first ones have bending strength of 690 N.mm^{-2} and the second ones - 930 N.mm^{-2}. The modulus of elasticity of these two composite types is respectively 70 000 and 53 000 N.mm^{-2} (Nabi Saheb and J.P. Jog, 1999). In this respect, the optimal bending strength can be achieved with the use of flax fibers and the best modulus of elasticity – with the use of hemp fibers.

Thermal properties of NFPCs depend mainly on the characteristics of the utilized natural fibers. In general, the thermal degradation process of NFPCs can be divided into the following five stages:1)100-200 °C - weight loss of NFPCs because of water evaporation from the fiber surfaces; 2) 200-270 °C - thermal decomposition of hemicelluloses; 250-350 °C - thermal decomposition of celluloses; 280-500 °C - thermal decomposition of lignin; 5) 200-500 °C - polymer matrix macromolecular degradation or depolymerization (Saheb and Jog, 1999; Monteiro et al., 2012; Stokke et al., 2014; Beyler and Hirschler, 2001). Degradation of the natural fibers will result in decreased mechanical properties as well as in changes in odour and colour of the composites (Gonzalez and Myers, 1993).

Water absorption of the natural fibers is typically from 7% to 20% (Stokke et al., 2014). The corresponding value for the polymer matrix materials is less than 0.9% (Holbery and Houston, 2006). In this respect, water absorption of NFPCs should increase with increased fiber content, which will have a negative effect on their mechanical properties. Water absorption of NFPCs is dependent to a great extent on fiber size. Fiber size has also significant influence on water absorption of the NFPC (Migneault et al., 2008). According to Tajvidi et al. (2006), the maximum water absorption of some natural fiber polymer composites, reinforced with different fibers (wood fibers, rice husks and kenaf fibers) is within the range 1.1–13.2%.

Despite the numerous advantages of natural fibers in the composition of the NFPCs, there are also some drawbacks, associated with excessive water absorption and poor thermal properties of these composites. In addition, there are some less studied aspects, such as thermal conductivity and acoustic insulation properties of these lignocellulosic composite materials. Future research on the properties of different natural fibers is required to obtain wider and safe use of these materials.

5. CONCLUSIONS

The objectives and targets set in the European legislation concerning waste management aimed at reducing the greenhouse gas emissions and environmental pollution (directly by decreasing emissions from landfills and indirectly by recycling materials), have been the main drivers to stimulate the development of innovations in recycling, to limit the use of landfilling, and to create incentives to change social attitude. If one industry's waste and residues become another's raw material, we can

implement the principles of the circular economy by eliminating waste and using natural resources in an efficient and sustainable way. The improved waste management also helps to reduce human health problems and avoid other negative impacts such as landscape deterioration due to landfilling, water and air pollution, as well as littering.
The new composite materials, utilizing organic waste and residues from agricultural and industrial processes, represent an important environmental alternative to the traditional composites. Due to their renewability and biodegradability, natural fibers have multiple advantages over the conventional synthetic fiber-reinforced thermoplastic materials and can be effectively used for various commercial applications.

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