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Biopolymers in wood-based materials – a recent review

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Abstract: *Biopolymers in wood-based materials – a recent review*. The aim of the paper was to summarize the current state-of-art in the field of biopolymers application in the composites based on lignocellulosic raw materials. The cited literature show, in research and experiments, how promising the green composites market becomes. Biocomposites are becoming more interesting and promising alternative to commonly used petropolymers, which have a negative impact on health and the environment.

Keywords: biopolymer, wood, composite, polylactic, polyhydroxybutyrate, polycaprolactone

INTRODUCTION

Products of biological origin, both from renewable sources and waste materials, have been appreciated more and more in the industries, market and the scientific community. It is because of growing concerns about the dominance of non-renewable products whose resources are limited. Researches focused on economic and environmental problems are being carried out around the world, trying to improve the use of renewable resources in the production of composite materials. Natural fibres can be classified by source of acquisition, such as animal fibres (wool, silk) and plant fibres: wood fibres (soft and hard wood, recycled) and non-wood fibres (kenaf, flax, jute, hemp, cotton, agave, sisal, wheat, corn, bamboo) (Mohanty *et al.* 2005).

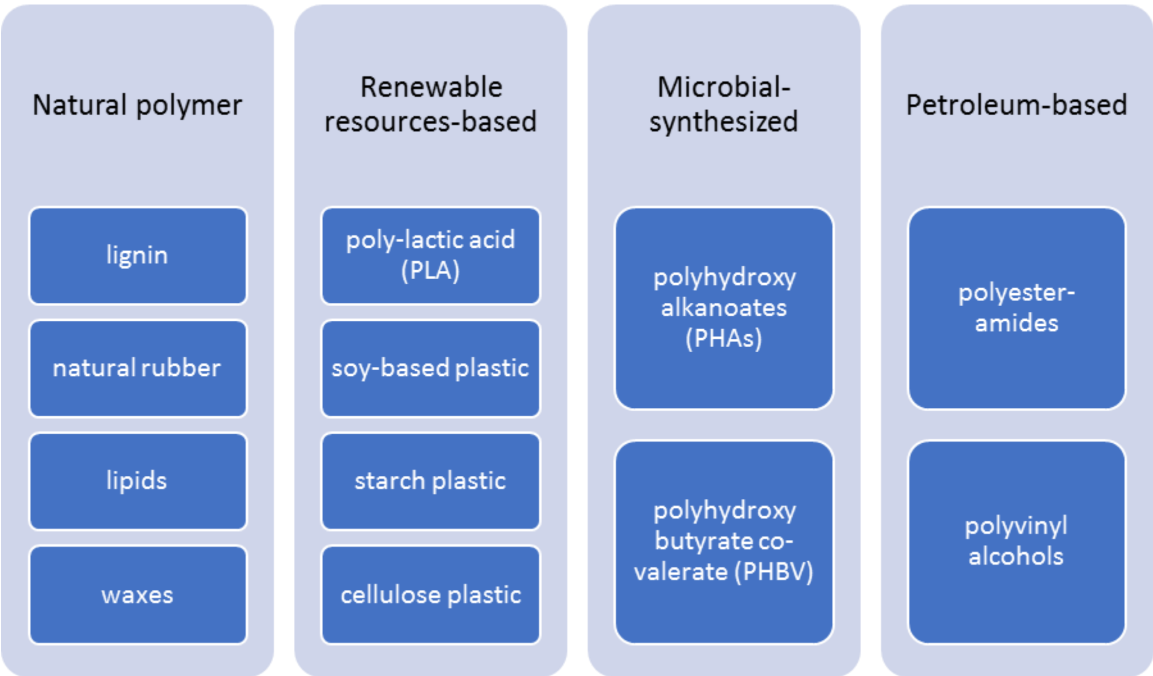

Figure 1. Classification of biopolymers (own elaboration based on Lau and Hung 2017)

Wood-based composites can be divided into 3 groups of panels, depending on the form of their structural components (fibrous, particle and sheet/layer). The structure of these panels and process engineering define their different mechanical and physical properties (González-García *et al.* 2009).

In case of lignocellulosic composites, their properties do not depend only on lignocellulosic material. Except for a few cases, these composites are generally produced with the use of bonding agents. Currently, these binders are based on depleting fossil fuel resources. Due to this, more and more attention is paid to adhesives produced from natural raw materials and by-products. Although these adhesives of vegetable or animal origin have been known almost as long as humanity exists, the expansion of science and knowledge of the chemistry, as well as improved and continuously developed technical formulations, are still necessary to intensify their broader industrial use. An intensive progress in the application of a wide range of biopolymers is observed, as well. Several of them (Figure 1) have been tested in terms of their application in wood-based composites

FIBROUS COMPOSITES

The first group from the entire branch of wood-based composites is a group of fibrous composites (including fibres in a matrix), such as Wood Plastic Composites (WPC), fibreboard produced in dry method (HDF – High Density Fibreboard, MDF – Medium Density Fibreboard) and fibreboard produced in wet method. In this group of composites, wood fibres belong to the dispersed phase, ensuring stiffness and strength, while the matrix used is a structural adhesive, and can be in the form of synthetic resin, biopolymer or inorganic compound (Dai and Fan 2013). Since 1950 to 2018, the total production of plastic composites reached 6.3 billion tonnes, of which 9% were recycled and 12% were incinerated. According to forecasts, this proportion will change in the future to a higher recycling and biobased polymers use ratio (Figure 2). Currently, the demand for plastic composites made from (petro-) polymers is higher than for composites based on biopolymers (Okunola *et al.* 2019). In 2015, inorganic matrices constituted 43.4% and natural polymer matrices were 25.1%. It is estimated that over the next few years, natural polymer matrices will have a relatively larger market share and demand, compared to synthetic polymers (Balla *et al.* 2019). In order to improve the adhesion between the matrix and the dispersed part, chemical treatments are used, e.g. fibre surface treatment 5% – NaOH; 0.5% – silane (Pickering *et al.* 2011), physical treatments, e.g. corona discharge (Ragoubi *et al.* 2012) and plasma treatment (Jang *et al.* 2012).

The production of wood-plastic composites (WPCs) from a thermoplastic matrix derived from renewable raw materials is very desirable field on the world market. Polyhydroxyalkanoates (PHAs), which include polyhydroxybutyrate (PHB), polylactic acid (PLA) and polycaprolactone (PCL), are seen as replacements for commonly used polyolefins (Chen *et al.* 2016; Nagarajan *et al.* 2016). Polyhydroxyalkanoates (PHA) are biopolyesters made from natural resources using various chemical polymerization methods, therefore they are completely renewable, biodegradable and environmentally friendly (Amache *et al.* 2013). They are a substitution for products, whose utilization has a negative impact on the environment, and they reduce the need for fuels and petroleum products (Bugnicourt *et al.* 2014).

The most common biopolymer – PLA (Castro-Aguirre *et al.* 2016) – and the above-mentioned polyhydroxyalkanoates have very high biocompatibility and are potentially biodegradable. They are widely used in the medical and packaging industries and attract attention of the wood-based panels industry. Pure PLA also has disadvantages, the main one is low ductility and toughness, which makes it not used in durable-requiring applications

(Nagarajan *et al.* 2016). In order to improve the mechanical properties of plastic composites, they are modified by introducing fillers and various additives (Wahit *et al.* 2012). The most common fillers include natural fibres because they are widely available and relatively inexpensive compared to synthetic fibres (e.g. glass fibres), which translates into lower costs of the final product (Awal *et al.* 2015, Dalu *et al.* 2019). The advantages of biocomposites with wood fibres are: low weight, formability and easy processing, and design attributes (Mertens *et al.* 2017).

Kuciel *et al.* (2020) produced biodegradable composites based on polylactic acid with basalt fibres (BFs) or wood fibres (WFs) with a PLA content of 7.5 or 15%. An analysis of the results confirms the correctness of using natural fibres as fillers. The mechanical properties and dimensional stability of composites have significantly improved. In one of the study, poor adhesion between PLA and thermoplastic starch (TPS) was increased by adding wood fibres (WFs), what improved mechanical properties of composite. Increasing the content WF up to 40% additionally increased the flexural (bending) strength by 180% and tensile strength by 128%, compared to a reference TPS/PLA blends without the participation of wood fibres (Raghu *et al.* 2018). Seggiani *et al.* (2015) produced blends with Polyethyleneglycol (PEG), in BHA matrix with and without wood fibres. Way *et al.* (2013), using up to 50% w/w of the maple wood fibres (WF) and up to 25% w/w of the waste cotton fibres (CL) in the PLA matrix, have observed an improvement in the stiffness and strength properties of the produced PLA-CL and PLA-WF composites. The increase in tensile strength and modulus of elasticity was 51% and 98%, respectively, and the increase in the value of the flexural strength and modulus of elasticity was 56% and 123%, in relation to pure PLA. Also, other researchers have explored the connection of wood fibres with PHA as a binder (Dimonie and Râpă 2010; Tănase *et al.* 2015; Csikós *et al.* 2015).

Fibreboards are manufactured by two methods: the dry method, where the fibre-carrying medium is air; and the wet method, in which water is the carrying medium. MDF panels are wood-based materials made by compressing lignocellulosic fibres (80% fibres in the final composite) with the addition of synthetic resins, under high pressure and temperature. Fibreboards show homogeneous density and raw material composition throughout their cross-section, thanks to which they have excellent machinability in the cutting process and is mainly used in the furniture industry. Medium density fibreboards production capacity is over 25 million tons per year around the world (http://bc.bangor.ac.uk/). Currently, in the fibreboard, particleboard and plywood industry, synthetic adhesives such as urea-formaldehyde (UF), melamine-urea-formaldehyde (MUF) and phenol-formaldehyde (PF) are widely used due to their desired efficiency and low costs of production, while offering favourable adhesive properties. Of course, they have the disadvantage of free formaldehyde emissions, which is a health and environmental problem (Salleh *et al.* 2015). Environmental regulations pressure wood-based panels producers to consider green technologies.

Many researchers have started looking for an environmentally friendly alternative to solve this problem. One of the ideas was to propose the production of binderless wood based panels, in which the self-bonding phenomenon of particles depends only on the content of chemical components in the lignocellulosic material. Components with adhesive properties include, i.e. lignin and polysaccharides (Boon *et al.* 2019). Domínguez-Robles *et al.* (2018) used self-lignin as a green adhesive in the production of HDF boards made of wheat straw. Lignin was obtained in the same process as cellulose fibres, so no additional steps were needed and the process was more efficient. The value of bonding strength for panels produced only from wheat straw fibres was higher than the value obtained for commercial HDF panels. In the tests they applied lignin content in the range of 0–15%. The w/w share of 15% lignin in the fibreboard proved to be the most advantageous (bending strength 96.81 MPa, modulus of elasticity 3550 MPa, internal bond 1.46 MPa). Such favourable results valorise straw, which is considered waste, left in the fields or burned, and thus the use of wood for the production of fibreboard could be decreased.

Zhou *et al.* (2013) tested the influence of oxygen plasma processing time and power on the mechanical and physical properties of composites from poplar fibres, and plasma-treated enzymatic hydrolysis lignin (EHL) was evaluated. They have determined that 5 min and 200 W of plasma treatment is sufficient for the application of EHL as a natural binder in producing high-performance lignocellulosic composites.

Corn stalk biomass and cellulose nanofibres (CNF) reinforcing agent were used to produce binderless fibreboard (Theng *et al.* 2015). It was found that increasing CNF content in the panel has a favourable effect on strength properties. Another alternative to synthetic adhesives is the application of natural and renewable source adhesives. Natural binders, including starch and its modifications (Tan *et al.* 2011; Gadhave *et al.* 2017; Wang *et al.* 2017), chitosan (Patel *et al.* 2013), tannins (Roffael *et al.* 2000), citric acid (Umemura *et al.* 2014).

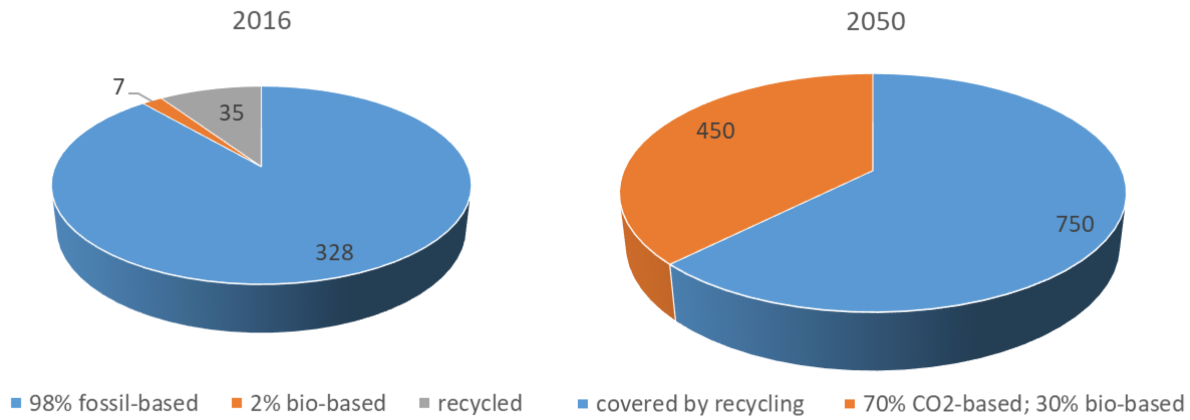

Figure 2. World Plastic Production and Carbon Feedstock in 2016 and Forecast for 2050 (in Million tonnes) (own elaboration based on "Renewable Carbon – Bio- and CO_2-based Economy" 2018)

Akbari *et al.* (2014) conducted investigations on natural rubber (NR), latex and starch as a binder to improve mechanical and physical properties in MDF boards. The panels were made in 3 combinations, then the results were compared with commercial MDF boards. They proved that NR latex and starch are a viable alternative to commonly used synthetic resins.

Battegazzore *et al.* (2019) presented an attempt to use agricultural waste and by-products (hemp fibres and rice husk particles) for the production of fire resistant fibreboards and particleboards, while they confirm the possibility of designing materials derived in 90% by weight from renewable sources.

PARTICLE-BASED MATERIALS

Depending on the form of their structural components, the second group of panels are composites made of particles (particles in the matrix), such as particleboards (PB), or the Oriented Strand Board (OSB). As mentioned above, commercial particleboards are manufactured based on amine resins. Small wood particles (e.g. sawdust, shavings, strips) and/or other lignocellulosic particles (e.g. flax, hemp, bagasse) are hot pressed under high pressure. In this group of wood-based panels, binderless particleboards were also tested as an alternative to synthetic adhesives. Baskaran *et al.* (2012) for the production of binderless boards, have used oil palm biomass and the addition of two variants of polyhydroxyalkanoate, freeze dried PHA and pure PHA into the panels. Composites with pure PHA resulted in better internal bond (IB) strength and modulus of rupture (MOR), compared to other panels. Thickness swelling (TS) and water absorption (WA) decreased with increased values of PHA.

Oil palm waste has been widely studied by Hashim *et al.* (2012). Oil palm biomass is a promising raw material for the production of this type of panels, due to the high content of starch and sugars (Nadhari *et al.* 2013). Baskaran *et al.* (2019) also produced particleboards from waste oil palm trunk, using PLA as a natural binder. The target board density was 800 kg/m^3, 10% PLA, thickness of the produced panels was variable: 5, 10 and 15 mm. A significant improvement in mechanical properties (MOR and IB) was noted and physical properties (TS and WA) significantly increased after the addition of PLA in relation to the binderless particleboard.

Nonaka *et al.* (2013) characterized binderless particleboard produced from sugarcane bagasse, in the range of high press temperatures (200–280°C). The reference particleboard was connected with diphenylmethylene diisocyanate resin (PMDI) at 200°C. The variant of particleboard from bagasse pressed at 260°C, reached the MOR value at the similar level of the reference board and the thickness swelling (TS) value was lower by 3.7% than the PMDI particleboard.

The use of modified starch as a natural binder in the production of particleboards has been extensively studied (Amini *et al.* 2013; Ye *et al.* 2018; Akinyemi *et al.* 2019). The development and testing of natural adhesives and their modifications showing good binding properties is still an industrial challenge.

LAYERED WOOD-BASED MATERIALS

The third group of lignocellulosic-based composites, named here layered composites, is plywood and LVL. These are wood materials produced as a result of press at high temperature, where layers (sheets) of wood (veneer) of small thickness will combine with amine or phenolic resins.

Luedtke *et al.* (2019) investigated the PLA combination – amorphous PLA (PLA 4060D) and semi-crystalline PLA (PLA3052D) with a wood surface bonding. Layered composites have been manufactured from two wood species, they used maple (*Acer pseudoplatanus*) and beech *(Fagus sylvatica)* veneers. Composites were pressed at 140–160°C and the variable was press temperature (45–100°C). When the press temperature increases, the viscosity of PLA decreases, giving rise to a better interferences with the wood structure. When analyzing the mechanical properties, no difference was observed between the PLA form used. However, considering the wood species, maple wood retained higher values for strength properties than beech wood.

Gaugler *et al.* (2019) used polymers as well as biopolymer, amorphous PLA (PLA 4060D) and semi-crystalline PLA (PLA3052D) to assess the interfacial adhesion, using the Automated Bonding Evaluation System (ABES) to allow rapid preparation (heating and cooling) and testing thermoplastic composites at various temperatures. For PLA-based composites, shear strength was affected by compression temperature. The shear strength increased as the compaction temperature increased.

Researchers, Bakken and Taleyarkhan (2020) have studied the use of two PLA polymer-based adhesive formulations based on amorphous (PLA-10361D) and crystalline (PLA-4043D-30C) polymers. Preparation and subsequent tests were performed on 2- and 3-layers of softwood plywood. Binder was applied in dry powder and wet spray modes. In the case of 2-layer plywood for both PLA variants, satisfactory strength and resistance to soaking were obtained. Tests for 3-layer plywood and crystalline PLA did not withstand 1 soaking cycle of samples (with 300 g/m^2 applied).

Another example of applying PLA and PHB as a natural matrix in layered composites have been studies by (Battegazzore *et al.* 2019), where cotton sheets were used. A 9-layer composite with a thickness of 4 mm was created by alternating layers of dry cotton fabric and a 0.2 mm-thick matrix film. The results of bending strength and modulus of elasticity were compared to particleboards (there are seven classes of use depending on the conditions – EN 312). All composites with PLA matrix meet the highest requirements of 22 MPa in terms of bending strength and 3350 MPa for the modulus of elasticity. PHB matrix composites showed good values for bending strength, but not for the modulus of elasticity in the highest requirements.

The current state of knowledge is limited on the subject of the use of polyhydroxyalkanoates (e.g. PLA, PHB) as a binder in wood-based panels technology. There is a lack of information about trials to apply the remaining biopolymers in wood-based composites, especially layered panels.

CONCLUSIONS

The research and experiments presented above show how promising the green composites market is. Biocomposites are becoming a more interesting and promising alternative to the commonly used petropolymers, which have a negative impact on health and the environment.

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Streszczenie: *Biopolimery w materiałach drewnopochodnych – najnowszy przegląd.* Celem pracy było podsumowanie aktualnego stanu wiedzy w zakresie zastosowania biopolimerów w kompozytach opartych na surowcach lignocelulozowych. Cytowana literatura pokazuje, w badaniach i eksperymentach, jak obiecujący staje się rynek „zielonych kompozytów". Biokompozyty stają się ciekawszą i obiecującą alternatywą dla powszechnie stosowanych petropolimerów, które mają negatywny wpływ na zdrowie i środowisko.

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