RESEARCH ARTICLE

Biotechnological methods in pulp and paper production from an alternative source of raw materials - the savior of the ecosystem

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Abstract: The pulp and paper industry is at its peak due to the annual growth in demand for paper products which includes packaging board, hygienic paper products, printing papers and the list is endless. To date, the main source of raw materials for pulp and paper production is wood. However, its use and the use of standard flow charts in pulp and paper production creates environmental problems. For example, deforestation to obtain raw materials leads to a disruption of biogeocenoses, and technology using alkalis and acids are sources of toxic wastewater that pollute the hydrosphere and lithosphere. In order to avoid the above environmental problems, other options for sustainable sources of raw materials can be considered, such as agricultural waste - straw. By its natural origin, straw is a non-woody plant, which will significantly simplify the flow chart of pulp and paper production, on the one hand, and on the other hand, the use of straw in production will help to reduce the risks of early global warming, since most of the straw is burned in the fields. As far as toxic wastewater emissions are concerned, they can be drastically reduced through the biological degradation of straw lignin to produce pure pulp. Biodegradation of lignin can be produced by widespread wood-destroying fungi due to their ability to synthesize lignin-degrading enzymes. Thus, the use of biological agents in pulp and paper production will make it possible to eliminate the use of caustic acids and alkalis.
Keywords: Biodegradation, catalysts, enzymes, fungi, lignin, mediators, non-waste production, toxic waste, wheat straw

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1 Introduction

With the growth of the Earth's population, industrialization and urbanization of modern society, the demands on the pulp and paper industry have also increased. Annual demand for paper products results in the loss of massive forest areas (Ervasti, 2016) . More than 390 million tons of paper and paperboard are currently produced worldwide (PG , 2018). In addition to stationery, newspapers and textbooks, the production of packaging and hygiene products is in greater demand (Berg and Lingqvist, 2019). Competition among suppliers of raw materials for pulp and paper production is certainly increasing, leading to massive deforestation, with much larger areas of deforestation than new plantations. Consequently, by reducing the area of forest plantations, the cost of wood is only increasing, which is another reason why it is necessary to introduce alternative affordable raw materials into the pulp and paper industry. To solve this problem, it is possible to use the waste of agriculture - straw, as an alternative raw material.

Straw is a common waste of agriculture, so the concept of waste-free production can be supported. There are three main sources of agricultural waste: plant stems, cob scales not used in food production; technical crops grown in agriculture and wild crops (Suseno et al., 2019). Previously, the waste was disposed of by incineration or used for composting. However, through the development of biotechnology, scientists have discovered the great potential of straw waste for biofuels and other forms of energy due to the rich component composition (Zhang, 2010, Otieno and Ogutu, 2020). Another advantage of straw as an alternative to wood raw material is excellent fiber content, for the production of special papers such as filter, kraft, vellum and newspaper (Aprianti, 2019, Suseno et al., 2019).

China and India are currently the world's leading producers of pulp products from alternative raw materials. It was found that China produces more than 70% of its paper products from non-wood raw materials such as: rice husks, corn cobs, etc., and India about 8% (Otieno and Ogutu, 2020).

In addition, agricultural residues are considered sustainable sources of raw materials, as they accumulate in agricultural fields after harvest, or even several times a year depending on the varieties of cereal plants.

2 Pulp and paper production worldwide

The pulp and paper industry is considered one of the largest industries worldwide. For example, paper consumption in China has increased dramatically in recent years, resulting in increased demand for pulp and paper raw materials (Zhao et al., 2010). Leaders in this industry are such countries as: North America, Northern Europe and East Asia. Australian and Latin American countries also make a significant contribution to the development of this industry. China and India provide pulp and paper products in Asia, together covering about 78% of all paper needs (Abd El-Sayed et al., 2020). The current annual consumption of pulp and paper products is 390 million tons (PG , 2018). China, the United States and Japan are the largest consumers of the industry, due to population, urbanization and industrial growth. The increase in demand for paper can be explained by the use of many industrial enterprises of paper packaging products and the demand for hygiene products such as: disposable paper towels, toilet paper and napkins for beauty (Johnston, 2016).

Any production is directly dependent on the raw materials, so for example, the paper production we consider depends on the availability of raw materials, which are wood forms, but due to the reduction in the number of forest plantations and the long recovery period, the industry requires the introduction of an alternative source of raw materials. One of the promising, stable and biodegradable alternative is straw - the waste of the agricultural sector. The use of non-wood plant species in papermaking was first practiced in Egypt when papyrus paper was made from papyrus sedge in 1800 BC (Kamoga et al., 2013). However, the decisive factor was the discovery by the French scientist Anselme Payen in 1833 that the non-wood forms of plants are the main sources of cellulose, so it is the main structural component of the cell wall of these plants. For the first time the USA has commercialized paper production from non-wood raw materials, after which this technology began to widely spread in other countries (Ogunwusi, 2014). To date, Malaysia uses palm waste as raw material for pulp and paper production due to the large amount of waste amounting to 23 million tons per year, and about 90% of them further recycled (Aljuboori, 2013, Padzil et al., 2020).

Some of the most common non-wood paper plants were cane pulp, hemp, kenaf and bamboo (Byrd et al., 2013). These plants have become popular for paper production due to their prevalence and accessibility. Another reason for the use of agricultural residues in the pulp and paper industry is that they contain the same fibers as hardwood (Zeeshan, 2011). The great potential for non-wood forms in their use is the possibility of producing special durable packaging materials by adding various chemicals and test fibers (Serna, 2017).

Non-wood plants belonging to the class of monocotyledonous plants such as sugar cane, maize have a similar fibrous composition to hardwood trees, which provides a good potential for their introduction into the pulp and paper industry. Today, the straw paper industry uses bleaching technology with chlorine, alkaline extraction and hypochlorite (Zhao et al., 2010).

3 Potential of straw for pulp and paper

A very important criterion when choosing a raw material is its availability and physical-chemical properties. Agricultural residues such as wheat straw are ideal raw materials for pulp and paper production, due to the high content of carbon compounds such as cellulose, hemicellulose (Harmsen and Huijgen, 2010, Tian et al., 2018). And also have a number of advantages over wood raw material: a) short maturation period or the stimulation of rapid ripening through the use of special fertilizers (Leponiemi, 2011); b) year-round accessibility due to the existence of spring and winter wheat varieties; c) low cost of raw materials, since straw is a by-product of agriculture.

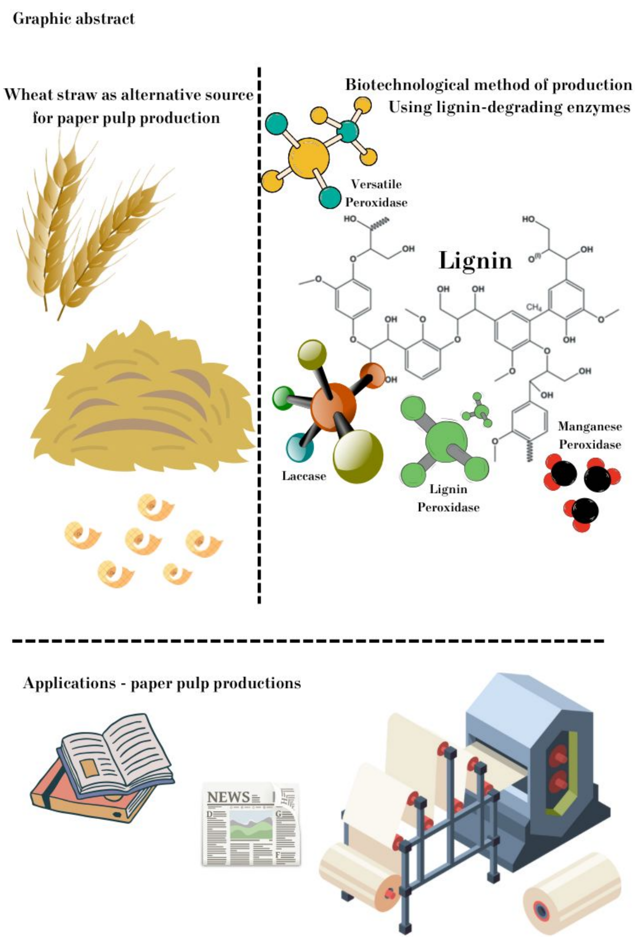

Figure 1. Flow chart of the use of straw and enzymes for pulp and paper production.

The concept of waste-free production implies the recycling of waste products into products with positive potential. Thus, the integrated management of affordable waste into usable finished products is relevant today. Many enterprises have as their main objective the introduction into production of waste-free production technology, or the development and implementation of technology that allows the use of waste from related industries (Gupta et al., 2020).

Currently, according to the official data of the Food and Agriculture Organization of the Unated Nations, 1.5 billion hectares are used for growing of agricultural crops (Food and Agriculture Organization of the United Nations, 2023). In the Republic of Kazakhstan, according to official data from the BNS ASP&R, the total updated area of agricultural crops for 2023 was 23.4 ml ha, which is a good indicator of agricultural development (Pryanikov, 2022).

World cereal production in 2023 was 2,773 million tons, of which 784 million tons were wheat. In the Republic of Kazakhstan in 2023, the gross grain yield of including pulses amounted to 18.6 thousand tons. That is 1% of the world production, of which the gross wheat harvest accounts for 13.7 thousand tons (Food and Agriculture Organization of the United Nations, 2023). Data on cereal cultivation in other countries can be found in Figure 2 below.

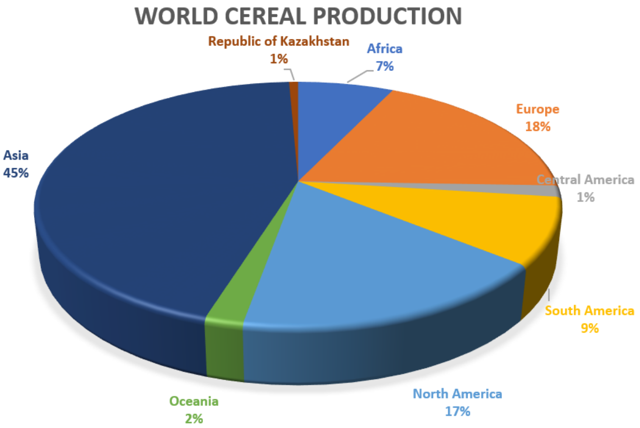

Figure 2. World cereal production.

In addition to the availability of raw materials, the transport of raw materials is of no less importance, for which there are also considerable financial costs, for example, due to the flexibility and small size of straw, more straw than wood can be packed in one package. It is worth noting and the number of trucks required for the transportation of raw materials is initially calculated based on the required amount of finished product. Another important factor in logistics is that many countries impose length limits on truck trailers (Peter and Hart, 2022).

The use of straw as a raw material for paper production leads to reduced or eliminating the burning of straw waste after harvest. In terms of waste quantities, an average of 750 million metric tons of straw are produced globally today (Tian et al., 2018). While the processing of 1 kg of wheat grain produces on average about 1.3-1.4 kg of wheat straw (Rencoret et al., 2012), which is a waste, yet has great potential in various production areas. But there is also the advantage of simplifying production technology and reducing the number of necessary reagents, which will reduce the cost of finished products. Counterarguments to the use of straw as a source of raw material for pulp and paper production are the rationalization of forest management to preserve natural biogeocenosis and maintain the equilibrium of flora and fauna.

4 Physical-Chemical Characteristics of Straw

The quality of the final product depends on the physical and chemical properties of the raw material. So, for example, in the case of paper great importance is the durability of the produced cellulose because the shelf life of paper products depends on it. Earlier studies on the durability of straw cellulose have shown that straw has the most stable fibers among other representatives of non-wood and woody forms (Hammett et al., 2001). The difference in chemical composition between wood and non-wood raw materials is shown in Table 1.

Table 1. Chemical composition of the straw and wood

	Cellulose	Lignin	Hemicellulose	References
Straw	32-47%	5-24%	19-27%	D'Souza, et al., 1996
Wood	40-45%	20-40%	15-30%	Rowell, et al., 2005 Fengel, et al., 1989

The presence of lignin in cellulosic raw materials has a negative impact on the strength and durability of the paper product. Table 1 shows that lignin is least present in non-wood plant forms, giving them an advantage. On the other hand, lignin has an antioxidant property, which plays a positive role in preventing the biological contamination of mold fungi and their enzymes (Barclay et al., 1997). However, numerous studies have shown that lignin is more susceptible to oxidation by the oxygen contained in the air, which can further cause paper oxidation. (Łojewski et al., 2010, Zervos, 2010). Figure 3 shows the structure of plant fiber consisting of cellulose, hemicellulose and lignin.

Thus, the main task of the preparatory stage of raw material processing is to reduce the amount of lignin in cellulosic raw materials to a minimum, in order to improve the quality of final products. Researchers have found that treatment of straw raw material with 2% (wt.) alkali solution allows to reduce the amount of lignin by almost half, due to the destruction of strong hydrogen bonds in the cellulose-hemicellulose structure, under the influence of the OH group of the used alkali (Chowdhury et al., 2013). Other scientists like Silvia Bolado-Rodriguez, Christina Toquero, Judith Martin-Juarez,

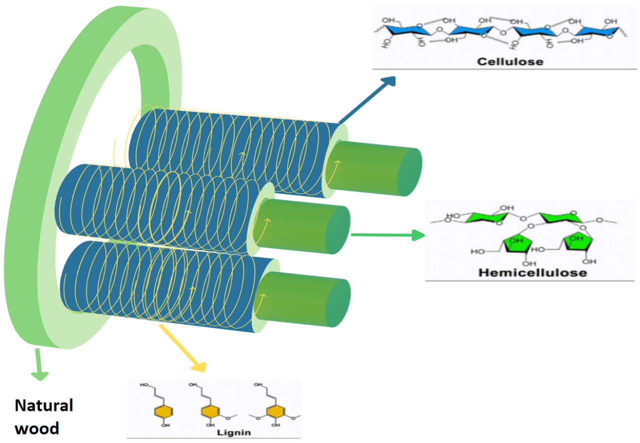

Figure 3. Plant fiber.

Rodolfo Travaini, Pedro Antonio Garcia-Encina found that the alkali dissolved lignin, forming acetic acid and phenolic compounds (Bolado-Rodrguez et al., 2016).

However, straw cellulose has well-ordered hydrogen bonds, which gives it a high density even after alkali treatment (Chowdhury et al., 2013), which will allow the production of dense paper with high physical-mechanical properties.

The morphological properties of straw also play an important role. For example, untreated straw compared to treated straw has a non-uniform loose rough surface, which allows reagents to penetrate more quickly into the internal structures of the straw (Haque et al., 2022).

Mechanical properties of raw materials - conventionally denote strength, extensibility of paper products. The straw raw material treated with alkalis and liginolytic enzymes has increased mechanical properties due to adhesion of heterogeneous bodies due to free active hydroxyl groups formed during leaching or biodegradation of lignin. In other words, the removal of lignin from the plant cell wall enhances the crystallization of the fibers, which gives tensile stability (Ray et al., 2004, Silva et al., 2017).

With the development of biotechnology, natural components have also been considered in terms of biodegradability. Thus, as mentioned earlier, straw has a looser heterogeneous structure, in addition to the absence of wood in the straw, which allows microorganisms to penetrate more actively and faster into plant cells. While wood raw materials for biodegradation require pre-treatment: grind, soak, to ensure looseness and easy permeability by microorganisms (Ringstrom, 2019).

Other studies have found that the final product made from straw fiber has equal or improved physical properties compared to the paper based on wood raw materials. To date, Kimberly-Clark has introduced a technology for the production of GreenHarvest napkins, which includes non-wood fibers of straw and bamboo pulp in the amount of 20%(Peter and Hart, 2022). Another example of the use of non-wood raw materials is Essity, fabric manufacturing company, which has patented a technology to produce fabrics with a non-wood fiber content of 20%(Ringstrom, 2019).

Thus, a physical-chemical comparison of wheat straw (FP) proved that FP could be used as one of the most promising alternatives to wood raw materials for the pulp and paper industry.

5 Biotechnological Method for Pulp and Paper Production

The paper production process, regardless of the raw material, consists of two main stages: a) preparation of the raw material and release of cellulose from the raw material for further use; b) production of paper using cellulose obtained during the preparation process.

Raw material preparation

Like any other production and paper mill starts at the preparation stage which includes pre-treatment of raw materials in the case of wood raw material (Bajpai, 2010) while the use of non-wood raw materials allows to skip this stage, because straw has no woody bark. The next stage is mechanical grinding to the required dimensions, according to the technology (Anuar et al., 2018). And the last stage of preparation is crushing, which takes place in special apparatuses called crushers, there the raw material is crushed to the condition of chips (Sharma et al., 2015).

Release of Cellulose

Plant tissue consists of three main structural components: cellulose, hemicellulose and lignin, but cellulose and hemicellulose are generally considered to be the major fibrous components. Since the target component of the pulp and paper industry is cellulose, hence the key is the stage of removing unnecessary lignin. In the conventional papermaking method, either a mechanical method with using high temperature and pressure explosions (Bajpai, 2010, Hart, 2011) or a chemical method with alkaline solutions (Ogunwusi and Ibrahim, 2014, May et al., 2015, Sharma et al., 2015) are used for the lignin separation process. However, these methods have a number of disadvantages: a) thermal emissions to the atmosphere (global warming); b) emissions of toxic solutions of alkalis into the ecosystem, causing pollution of groundwater and soil.

The three most commonly used methods for releasing the main fibrous components are mechanical, chemical or semichemical and biological. The alternative is a biotechnological method - the biodegradation of lignin by microorganisms and/or by the ligninolytic enzymes that synthesize them, the advantage being that the action of the enzymes can be controlled. To release cellulose, the crushed chips are placed in the bioreactor, and cultural liquid with lignin-degrading abilities is introduced, the duration of the delignification process for each microorganism is individual. In this case, it is also worth considering the use of mediators and catalysts that affect the process of biodelignification. After the required time has elapsed, the cellulose mass can be extracted from the culture liquid and the residues of the plant cell.

In addition to separating lignin from cellulose, the biological treatment method has the advantage of reducing lipophilic extracts in cellulose (Pandey et al., 2022). To date, there has been much scientific work on the detection of microorganisms capable of destroying wood, including fungi that cause brown white rot on the surface of rotting wood (Geles, 2007, Semenkova, 2008). Below is a table with the classification of fungi and the formation of the corresponding rot.

Table 2. Classification of fungi and their lignin degrading enzymes

Type of rot	Enzymes	References
White rot	LiP (EC1.11.1.14) MnP (EC 1.11.1.13) VP (EC 1.11.1.16)	Chen, et al., 2012 Dinis, et al., 2009 Elisashvili, et al., 2009
Brown rot	Laccase	D'Souza, et al., 1996.

The mechanism of lignin decomposition occurs due to the lignolytic enzymes complex of wood-destroying fungi and bacteria.

The most common lignin-degrading enzymes found in nature are: manganese peroxidase (MnP), lignin peroxidase (LiP) and laccase. Each enzyme has its own physical and chemical properties.

The lignin peroxidase is the first enzyme found in *Phanerochaete chrysosporium* with a high potential for lignin degradation, more precisely, it has a high potential to oxidize parts of the aromatic ring of the non-phenolic part of lignin, which form about 90% of the total polymer (Tien and Kirk, 1984).

Manganese peroxidase is also an extracellular enzyme capable of degrading lignin, one of the distinctive features of this enzyme is that it is dependent on Mn^{3+}, which acts as a charge carrier, and unlike lignin peroxidase does not have oxidation properties of the non-phenolic parts of lignin (Jeffries and Viikari, 1996). Catalysis by this enzyme begins with the coupling of hydrogen peroxide (H_2O_2) to the ferric enzyme and the formation of an iron peroxide complex (Hofrichter, 2002).

Laccas enzyme plays an important role in the process of lignin biodegradation. Laccases are copper-dependent oxidases that catalyses the single-election oxidation of four equivalents of the reducing substrate, with reduction of atmospheric oxygen to water (Dwivedi et al., 2011). Organisms capable of synthesising this enzyme are basidiomycetes, ascomycetes and deuteromycetes (Dedeyan et al., 2000).

Enzymes have a great influence on lignin degradation and today the actual task of biotechnology is to develop methods to stimulate the synthesis of lignin-degrading enzymes by microorganisms. Processing of raw materials with the potential to introduce these enzymes into the production process of pulp and paper production, or in the treatment of wastewater containing lignin. Another challenge is to identify new microorganisms capable of synthesising lignin-degrading enzymes or to breed new strains of microorganisms using genetic engineering.

Pulp processing stage

Pulp processing is most commonly used to remove contamination by sieving, declamation and defibrinating (Wan Daud and Law, 2010). Washing method is used to remove chemical residues (Rafidah et al., 2017). Washing not only provides purifies the pulp, but also makes it possible to return to production the culture liquids used in biodelignification, chemical reagents used in chemical pulping, for example. The most common equipment used during this stage is a rotary vacuum washer (Bajpai, 2010). Another equally important step is drying, this stage provides microbiological safety, as high humidity can contribute to the growth of yeast and fungi.

Pulp bleaching

The stage where the yellowness of the pulp is removed until a lighter tint is called the pulp bleaching stage. As a rule, unbleached pulp is acceptable for the production of kraft packaging materials, cardboard boxes. Thus the pulp bleaching process achieves two goals: to obtain a more presentable paper for the consumer, and to improve the chemical composition by removing the residual lignin, which to some extent gives yellow colour to the pulp. In total, about 5-10% of residual lignin is washed out during bleaching (Gopal et al., 2019). The pulp bleaching process can be carried out by different methods such as: chemical bleaching, which involves the use of chlorine and alkali (Latha et al., 2018).

The second method is biological bleaching using laccase and xylanase enzymes, thus eliminating the use of chlorine and alkali (Bajpai, 2010). Pulp produced by mechanical or semi-chemical pulping requires special attention, as the highest amount of lignin is observed in the pulp after these methods compared to the other methods.

After all stages of processing, 60-70% of the pulp mass consists of water, so the next stage is drying. The drying process takes place by evaporating the moisture using a drying machine (Eu and Nr, 2020). Drying is stopped when moisture in pulp is reached 2-8% of the mass (Bajpai et al., 2004). Dried cellulose is then grindable, wrapped in rolls and sent to storage.

The production of pulp and paper using straw as a raw material is generally similar to typical pulp production from wood raw materials.

During pulp processing, certain reactions take place to dissolve lignin or break cellulose-lignin bonds, and this lignin makes up the bulk of the black liquor. In this case, we get basic fibres with a small amount of lignin, which will be removed during bleaching.

Authors such as Pratima Bajpai, Shree Prakash Mishra, Om Prakash Mishra, Sanjay Kumar, Pramod K. and Sarju Singh studied the biological treatment of wheat straw by lignin-degrading fungi to study their influence on chemical processing of lignin. The use of *Ceriporiopsis subvermispora* reduced lignin content by 16.5%, and obtained cellulose had high brightness compared to the cellulose produced by the chemical method. Scientists also note that the content of toxic chemicals in wastewater was significantly reduced (Bajpai et al., 2004).

6 Mediators

Comparative analyses of plant cell size and lignin-degrading enzymes shown that enzymes are unable to penetrate the plant cell cavity. However, the use of natural enzymes in cellulose bleaching and lignin separation in pulp and paper production has great potential, but only with the use of madiators.

Mediators are low molecular compounds that have the ability to oxidize laccase. Due to their size, mediators are able to easily penetrate into the substrate and deliver enzyme molecules, while the enzyme due to its size cannot penetrate the substrate (Li et al., 1999).

An ideal mediator should have the following qualities: inexpensive, non-toxic, easily accessible, not inhibit enzyme action and have a stable redox potential (Morozova et al., 2007). Such mediators as 2,2'-azino-bis (3-ethylbenzothiazoline-6-sulphonic acid) (ABTS) and hydroxybenzotriazole (HBT) may even oxidize non-phenolic aromatic targets. Oxireductases require the use of low-molecular oxidizers such as veratryl alcohol, saturated lipids, or Mn2+ ions as redox mediators for lignin biodegradation (Martínez et al., 2018). To date, scientists are working to identify natural mediators, and suggest that phenolic derivatives of lignin, derived from the degradation of lignin itself, may be ideal mediators in lignin biodegradation (Camarero et al., 2007).

The mechanism of the mediators can be divided into two stages. First, the mediator enters into an oxidative reaction with an enzyme to form strong intermediates with high redox potential. The oxidized mediator then separates from the enzyme and penetrates into the plant cell due to its small size through the pores of the cell wall (Li et al., 1999). The primitive mediator for manganese peroxidase can be reduced Mn^{3+} which has the ability to oxidize the enzyme center (Kuan et al., 1993). The ideal mediator for lignin peroxidase is veratryl alcohol (Candeias and Harvey, 1995). Therefore, mediators in the process of lignin biodegradation are important components of the ligninolytic enzyme system. Mediators help enzymes such as MnP and LiP (Bourbonnais and Paice Michael, 1992) to reach native lignin present in wood (Datta et al., 2017).

7 Catalysts

Lignin biodegradation is based on the growth of microorganisms and their synthesis of lignin-destroying enzymes, but these microorganisms are often characterised by slow growth and lignin degrades much more slowly than other plant cell components. However, the addition of 0.12% of nitrogen to the culture medium of white rot fungi can accelerate the lignin degradation process at 39-40°C incubation from 5.2% to 29.8% (Yang et al., 1980).

Other scientists Loghavi et.al., Thrash, J.C., and Coates, J.D. believe that in order to improve lignin biodegradation process it is necessary to accelerate the growth of microorganisms on the substrate and propose the use of electrochemical technology (Thrash and Coates, 2008, Loghavi et al., 2009). It has been shown experimentally that the mass of the mycelium increases to 2.3 grams within 96 hours at a voltage of 0.3 A. In addition, it was found that under the influence of electron charges and Fe released by enzymes, oxidation reactions occur and hydroxyl groups are produced, which leads to lignin oxidation and easier degradation by enzymes (Shen et al., 2020).

As a catalyst is also worth mentioning the hydrogen peroxide (H_2O_2) that is required for the prosthetic iron group such as lignin peroxidase, to catalyze the oxidation of non-phenolic fragments of lignin. Manganese compounds are required for manganese peroxidase synthesis by wood-destroying fungi, in addition to the catalyzing function, Mn^{2+} acts as a mediator, being oxidized to Mn^{3+} during the lignin biodegradation process (Wariishi et al., 1989).

Thus the use of catalysts of chemical nature accelerates the process of lignin decomposition, due to oxidation and further detachment of phenolic groups of lignin.

8 Conclusion

Global paper production has now reached a record high of over 400 million tons, with most of the production of sanitary products and the demand for paper products growing daily. Due to the large number of orders, production is increasingly stocking up on raw materials, particularly wood products. But it is worth mentioning that every year the area of forest plantations decreases by an average of 6-7 million hectares, which has a directly proportional impact on fauna and the ecosystem as a whole.

Agricultural residues, such as grains, wheat, can be an ideal alternative for pulp and paper production. Using wheat straw can not only solve the problem of deforestation, but also slow the process of global warming, as most of the agricultural waste is burned in the fields without finding further use.

Any other production faces another problem, like wastewater. In this case, biotechnological production methods will minimise toxic emissions. Specifically, the replacement of acid-alkaline reagents to biologically synthesized lignin-degrading enzymes during puling. An extremely important advantage is the ability of inducing the synthesis of the necessary enzymes by wood-destroying fungi or bacteria.

This work shows that the production of cellulose from an alternative source of raw material - straw, using biotechnological methods, will save the area of planted forest plantation, will create a cycle of waste-free production between farmer cooperatives and industries, and the introduction of biotechnological methods in the technology map will reduce toxic emissions to the ecosystem.

Conflict of Interest

The authors declare no competing interests.

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