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Biodegradabiity of Non-wood Packaging Paper

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Abstract

The global packaging market is undergoing rapid transformation. The amount of package consumption is increasing, the market structure is shifting, and new packaging technologies are emerging. Biodegradable packaging is a solution to environmental challenges that is gaining increasing attention from the international community. The development of packaging that does not affect the environment in the process of disposal is presently a top priority. The process for making biodegradable packaging from natural biomass, namely agricultural waste and solid home waste, wollastonite, is presented in this article. α-cellulose, lignin, ash, and hemicellulose were examined for chemical composition in packaging paper made from agricultural waste, specifically wheat straw and rice straw, using established procedures. In addition, studies were conducted on the transmission of micro- and nanoparticles through the prepared paper, a study was conducted on an FTIR instrument to obtain cellulose spectra, and a thermogravimetric investigation to determine whether the samples were degradable, as well as a non-standard method for the biodegradable properties of paper.

Keywords: packaging, Biodegradable, natural biomass, thermogravimetric.

Introduction

Packaging is now widely used, and it should not only maintain food safety but also be economically feasible and environmentally friendly [1]. Production of biodegradable packaging from natural materials, which lowers the adverse impacts of different wastes on the environment, is one of the possible solutions to these challenges [2]. Because packaging is such a crucial component in the process of ensuring food quality, the food industry has very high regulations for packaging materials [3]. More than 67 million tons of packaging waste is generated annually in the world, which is about one-third of all municipal solid waste [4]. Every year in Kazakhstan, up to 6 million tons of solid domestic waste (MSW) accumulates, 30% is paper and cardboard, of which only about 11% is recycled, the rest is stored in landfills (Each Kazakhstani has more than 165 kg of garbage annually) [5]. Garbage disposal leads to an environmental problem for the country. In this regard, the Republic of Kazakhstan is planning measures to implement the concept for the transition to a "*Green Economy*" for 2021 - 2030 [6]. There have been several scientific discoveries in the field of new materials in developing countries with substantial agricultural production. It demonstrates the feasibility of using a resource-efficient method for agricultural waste processing to produce useful goods [7-9]. Because agricultural waste is burnt in many countries, harming the environment, there is a lot of interest all over the globe in inexpensive, yearly renewable plant raw materials as a source of fibrous materials [10-12]. Biodegradable packaging materials made from renewable natural resources have received a lot of attention in recent years all around the world. Biodegradable packaging helps to reduce the amount of landfills by using renewable and perhaps more sustainable raw materials. Agricultural waste, such as

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wheat and rice straw, is being investigated as a replacement for oil and gas in the pulp and paper industry. The fundamental rationale for picking wheat and rice straw is because we extract cellulose with a quality comparable to a wood pulp after delignification from straw [13] and it is inexpensive because it is an agricultural waste. Also, because of its unique properties - biodegradation - straw cellulose is one of the most significant materials [14,15]. In recent years, several academics have been interested in the bio-based in the form of straw as a renewable resource. For various sorts of items, several researchers have used bio-based packaging [16]. Agricultural waste packing paper, on the other hand, is of poor quality because it lacks fillers. Lamination with polyethylene, polypropylene, surface treatment with paraffin, adhesive formulations, and other materials are used to improve the quality of pulp and paper manufacturing and increase their application possibilities [17]. However, because the synthetic polymers in the materials' composition are practically not digested by the soil microbes, the biodegradability of the materials reduces [18]. A modified starch surface treatment was applied to increase the biodegradability qualities of the packaging paper. In most forms of paper, starch is a polysaccharide. Straw can be an excellent choice as a raw material for biological packaging from an economic standpoint. The higher biodegradability of bio-based packaging is a significant benefit in terms of product end-of-life waste management [19]. The goal of this research is to look into the possibility of using agricultural waste in the pulp and paper industry to make environmentally friendly packaging and high-demand products by looking into the chemical properties of cellulose, as well as the physical properties of paper using FTIR spectroscopy, thermogravimetric analysis, and biodegradable properties of paper.

2. Experimental

2.1 Materials

Wheat and rice straws from the Republic of Kazakhstan's (Akmola and Kyzylorda districts) harvests of 2019 and 2019 were utilized in this study. We also used the garbage from the home and second-hand items (cardboard). In a ratio of 35% : 35% : 23%, paper samples were made from shredded wheat and rice straws, as well as cardboard. Wollastonite powder (2% dry weight of raw ingredients), starch (4%), and rosin glue (1%).

2.2 Paper preparation

Using catalysts and continual stirring in a container with blades, shredded wheat, and rice straws, as well as cardboard, were cooked in an equilibrium peracetic acid ($CH_3COOH – H_2O_2 – H_2O$ – 2% H_2SO_4). After that, the samples were rinsed in distilled water until they reached neutral pH levels. The resultant pulp was poured with a 0.1 M sodium hydroxide solution and heated constantly stirring. After the container was processed, the impregnating solution was extracted. After that, 100 mL of water was added to the remaining suspension, and the cooking was resumed. The product was filtered and rinsed in distilled water until the pH level was neutral. The fibrous product was then heated with glacial acetic acid and hydrogen peroxide in the presence of a stabilizer to bleach it using the oxidative organosolvent method. After the cooking process, cellulose was treated with modified wollastonite [20] and modified starch. After mixing the suspension, the paper sheets were prepared by pressing them down with a hydraulic press and leveling them with a hand roller. The resulting paper sheets were then allowed to air dry at room temperature. The created paper sample was treated with a binder, potato starch, to bind the fibers together and generate a robust sheet of paper that may decompose later. The mofdified wollastonite from the Verkhnebadamskoye deposit in the Republic of Kazakhstan was utilized as a filler in the manufacturing of high-strength paper. The employment of this mineral filler in papermaking is determined by its strengthening capabilities, minimal water absorption, and chemical purity. Surface roughness was achieved by treating wollastonite in an acidic atmosphere. In terms of dispersibility, surface treatment is critical [20]. Table 1 shows the chemical composition of raw wollastonite from the Verkhnebadamskoye deposit. Wollastonite (49.4%), calcite (20.6%), quartz (20.2%), and pomegranates are found in the Verkhnebadamskoye deposit's wolostonite ore (5.1%).

2.3 Characterization of cellulose

The following indicators were used to determine the chemical composition of the cellulose obtained from straw: α-cellulose content according to GOST 6840-78, lignin content according to GOST 11960-79, and ash content GOST 18461-93.

2.4 Characterization of paper

Transmission of micro- and nanoparticles

Using a TSI 3785 particle counter and TSI VOAG 3450 aerosol generator, the transmission of micro-and nanoparticles with diameters of 0.015 – 0.30 microns, 0.3 – 0.50 microns, and 0.5 – 0.80 microns in air through the resultant paper and industrial wrapping

paper was examined. The ambient temperature was 21.2°C, relative humidity was 31.3%, and air pressure was 961 kPa. State meter of dispersion characteristics of aerosols of suspensions and powder materials, KZ.01.01.00049 – 2008 The measurement range for particle mass concentration is 1 to 1014 particles/m^3 and 0 to 2000 mg/m^3, with a 6% increased uncertainty.

Table 1. Chemical composition of wollastonite of the Verkhnebadamskoye deposit

Chemical composition	SiO_2	CaO	Al_2O_3	MgO	Fe_2O_3	T_iO_i	K_2O	Na_2O	MnO	n.nn	Total
Oxide content,%	36.51	48.41	2.62	3.23	3.62	0.04	0.78	1.12	0.3	0.05	96.68

FTIR spectroscopy

An FT-801 infrared spectrophotometer FTIR spectrometer was used to get the spectra of the final packing paper. The spectra were obtained with a resolution of 1 cm^{-1} in the spectral range of 500 – 4000 cm^{-1}. To make a comparison, we used commercial wrapping paper.

Thermogravimetric Analysis (TGA)

The thermal stability and fracture characteristics of the fibers in the resultant packaging paper were investigated using thermogravimetric analysis (TGA). The experiment was conducted in a nitrogen purge (N_2) environment with a TA Instrument Q-50 TGA thermal analyzer at temperatures ranging from 0 to 600°C. Data were analyzed using the TA Universal Analysis 2000 program. As a purge gas, nitrogen gas was employed, with a flow rate of 50 mL/min. The TGA Q50 V20.13 Build 39 instrument was used to determine how much weight was lost in the paper samples. The ASTM E1131-08 standard test technique for compositional analysis was used to carry out this procedure. Industrial wrapping paper was used as a baseline for comparison.

Differential Scanning Calorimetry (DSC)

Glass transition temperature (T_g) of hydrogels was tested by differential scanning calorimetry (DSC), on a NETZSCH, DSC200 PC, aluminum crimped pans under N_2 flow at 20 mL min^{-1} was used. The temperatures were carried out between 0 0C and 600°C at a heating rate of 10°C min^{-1}).

Biodegradability

The biodegradability of the samples (obtained paper; industrial wrapping paper) was assessed using a non-standard approach [21], employing non-sterile soil, as described by Li et al. [22,23]. The biodegradability of samples of 5 cm x 5 cm thickness were tested by placing them in a box with non-sterile soil and covering them with non-sterile wet soil, 25 – 30% moist soil. For ten weeks, the samples were maintained at room temperature. The examination was conducted out using the samples' visual and weight loss criteria.

3. Results and Discussion

3.1 Characterization of cellulose

The alkaline treatment causes the fibers to expand and the straw to loosen. The loosened straw is now ready for additional interactions with the components of the cooking liquor and successful delignification. Straw is made up of cellulose, hemicelluloses, and lignin and is a waste product from agricultural wastes [24]. Standard techniques of analysis were used to evaluate the chemical makeup of the acquired cellulose samples (mass fractions of cellulose according to α-cellulose, lignin). Table 2 shows the chemical characteristics of components before and after the chemical treatment of straw.

According to a study of ash and lignin content in rice (8.4% and 5.5%, respectively) and wheat straws (2.7% and 3.9%, respectively), wheat straw is the best choice for cellulose production [25]. The application of an additional step of organosolvent boiling resulted in the creation of purer fibrous products; for example, the lignin concentration in rice straw was decreased by about 4 – 5 times, from 21.3 to 5.5%, and in wheat straw from 21.2 to 3.9%. The fibrous components in wheat straw had the lowest ash concentration, at only 2.7%. The adoption of the three-stage process of wheat and rice straw delignification resulted in a high cellulose yield for paper manufacturing, according to the results of the analysis. Cereal straw contains more than 50% cellulose in plant materials [26,27], making it a viable cellulose source. The high amount of pentosans and ash in wheat and rice straw distinguishes them [28].

Table 2. Chemical parameters of components before and after chemical treatment of straw

Straw after processing	Component content			
	α-cellulose, %	Lignin, %	Ash, %	Hemicellulose, %
Rice straw	38.4	21.3	12.7	26.7
After all treatments	72.7	5.5	8.4	8.9
Wheat straw	44.5	21.2	4.3	24.5
After all treatments	76.2	3.9	2.7	12.3

The breaking of glycosidic linkages in amorphous regions is caused by pretreatment with acidic hydrolysis. During hydrolysis, amorphous hemicellulose decomposes to sugar monomers, as opposed to mechanical degradation, which destroys cellulose fibers in crystalline areas as well. After the hemicellulose is removed, the surface area rises and the pores expand, allowing for enzymatic treatment. Alkaline treatment hydrolyzes polysaccharide-lignin linkages, eliminates lignin, and reduces cellulose crystallinity [29]. Mineral components and a portion of lignin are eliminated when treated with an alkali solution, which contributes to the loosening of the ligno-carbohydrate material's structure and leads to the hydrolysis of the cellulose matrix [30]. Some non-cellulose components are eliminated from cellulose during hydrolysis, resulting in cellulose concentrations of up to 70%. The essential properties of the cellulose samples derived from wheat straw and rice are identical. It works by loosening and breaking the bonds in the cellulose fiber. When cellulose is ground, access to hydroxyls is enhanced, causing cellulose fibers to expand. Swelling contributes to the intensification of the process of bond breaking or weakening inside the fiber. The inclusion of hydroxyls from the contacting fibers in the water-hydrogen bond, as well as in the drying and hydrogen bond, contributes to an increase in the contact area of the fibers during the formation of paper and the formation of interfiber bonds due to the development of plasticity and flexibility of fibers as a result of weakening their internal structure.

The kinetics of the oxidative-organic-solvent boiling process were computed to elucidate the influence of acetic acid and hydrogen peroxide on the delignification process. The data are presented in Figure 1.

The presence of lignin is determined by the shift in acetic acid and hydrogen peroxide concentrations, as shown in the lignocarbohydrate matrix. Peroxy chemicals fully engage with the ligno-carbohydrate complex during the first stage of cooking, followed by lignin macromolecule fragmentation and the transport of water-soluble pieces into solution.

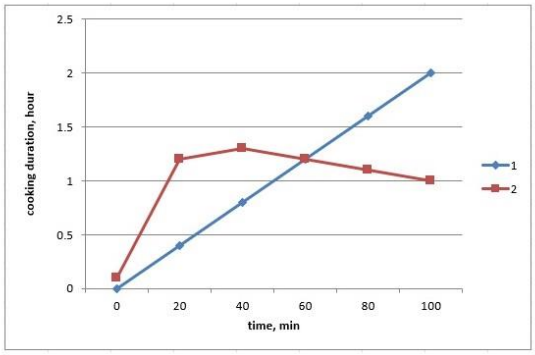

Fig. 1. Calculation of the kinetics of the process of oxidative-organosolvent cooking.

3.2 Characterization of paper

The number of micro-and nanoparticles in the air, as determined by the number of particles of various sizes in the air before and after passing through the paper of various compositions, were used to determine the throughput qualities of straw-based paper: (1) straw-and-cardboard-wrapped wrapping paper; (2) industrial wrapping paper (Figure 2). The results of the experiments showed that the generated paper samples might be used for a variety of functions, including filters and food packaging materials. Before using paper, the concentration of particles (n/cm^3) in the air with a size of 0.015 – 0.3 microns was 2435.587 (n/cm^3), with a size of 0.3 to 0.5 microns - 9.1 (n/cm^3) and from 0.5 to 0.8 μm - 0.983333 (n/cm^3). The concentration at the output for particles with a diameter of 0.015 – 0.3 μm was 78.85485 (n/cm^3), for particles with a diameter of 0.3 – 0.5 μm - 0.0465 (n/cm^3), and for particles with a diameter of 0.5 – 0.8 microns - 0.0465 (n/cm^3) after inserting the produced paper interlayer into the device.

Thus, when particles with a diameter of 0.015 – 0.3 microns were considered, air purification effectiveness was 96.76%, 99.19% for particles with a diameter of 0.3 – 0.5 microns, and 99.98% for particles with a diameter of 0.5 – 0.8 μm. Sample 1 had the highest micro- and nanoparticle transmittance, according to the study's findings. The starch binder, which is used to improve the adhesion forces between fibers and raise the strength of the paper in order to

produce a wide range of paper goods, played a significant role. One of the earliest and most widely used excipients in the manufacture of paper and paperboard is starch. Starch in the stock minimizes paper dust and improves filler retention. Furthermore, the paper's overall strength has been enhanced. The paper becomes stiffer and more elastic.

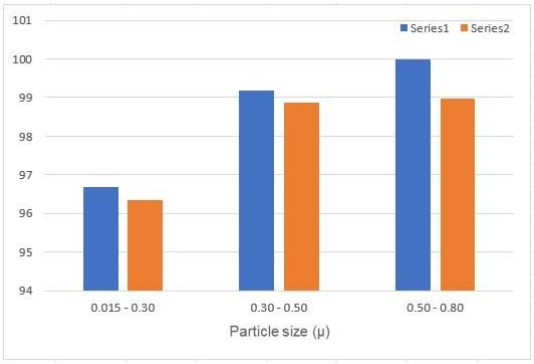

Fig. 2. Studies of paper samples for quantitative indicators of small particles with sizes of 0.015 – 0.3 microns, 0.3 –0.5 microns, and 0.5 – 0.8 microns in air.

The physicochemical parameters of the following materials were investigated using FTIR spectroscopy: straw-and-cardboard wrapping paper (A); industrial wrapping paper (B).

Fiber spectra for functional groups of wheat and rice straw cellulose are identical, with the predominant peak at 3431 cm^{-1} which attributed to stretching vibrations of OH, and the bands at 2989 cm^{-1} and 2817 cm^{-1} were associated with the vibration of the asymmetric and symmetric C-H group, respectively [30, 31], stretching and bending vibrations of 1123 cm^{-1} C-O and 904 cm^{-1} C-O-C of the cellulose ring, respectively [30, 32]. The presence of adsorbed moisture is demonstrated by broad absorption bands ranging from 3075 to 3783 cm^{-1}.

A strong signal in fibers at 1123 cm^{-1} suggests absorbed water molecules linked to cellulose fibers [31, 33]. The vibrations associated with the -C-O-H group have maxima in an absorption band of 1060 cm^{-1}. Peaks at 1620 cm^{-1} in the spectra show the presence of aldehyde and ketone groups of acetate groups in hemicellulose. The vibrations of the aromatic structures present in lignin may be responsible for the peaks in the 1445 – 1450 cm^{-1} area.

Thermal analysis (TA)

Thermal analysis is crucial in many sectors, including polymers, composites, medicines, foods, petroleum, inorganic and organic compounds, and many more. The thermal analysis (TA) behavior of the materials was investigated through experiments. The following samples were used: (1): straw-and-cardboard-wrapped wrapping paper; (2): industrial wrapping paper (2). Figure 4 shows the TGA and differential scanning calorimetry (DSC) findings of straw-based paper with and without wollastonite.

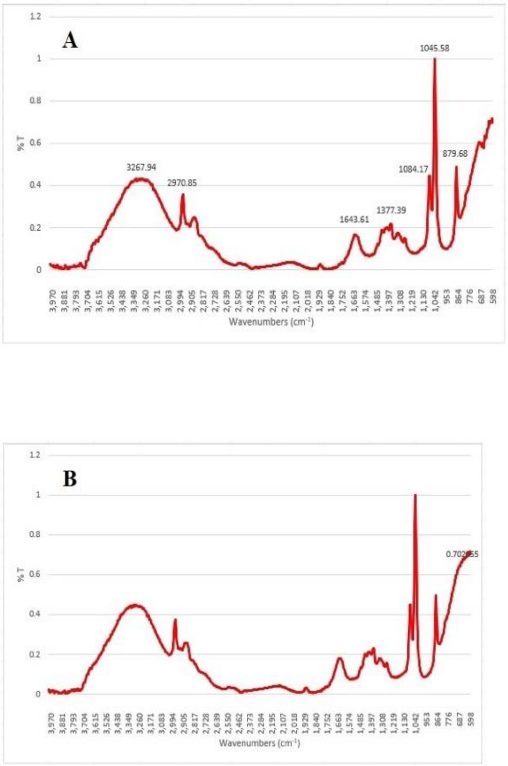

Fig. 3. FT-IR spectra of paper samples. Straw-and-cardboard wrapping paper (A); industrial wrapping paper (B).

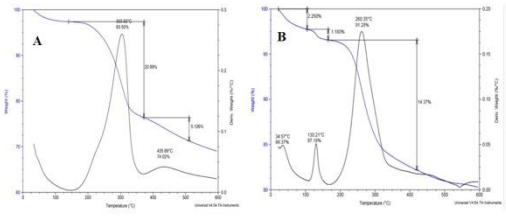

Fig. 4. TGA (A) and DSC (B) curves of paper samples.

The chemical structure of cellulose and natural wollastonite material differed somewhat, resulting in slightly differing thermal stability. Because cellulose degrades fast and constantly in this temperature range owing to the loss of hydrated water molecules from the

environment [34], both materials began to lose weight approximately 210°C. The TGA curve shows that paper with wollastonite loses weight at 303 and 435°C, but paper without wollastonite loses weight at 260 and 385°C. This is due to the loss of hydrated water molecules from the environment. The crystalline structure of cellulose probably generated lower breakdown temperatures than wollastonite paper, resulting in weight loss of around 260°C. Thermal degradation of the cellulose in the composite is likely to blame for the wollastonite paper's weight loss of roughly 303°C. With a weight loss of roughly 435°C in wollastonite paper, it can be connected with mineral filler oxides by conversion to β-wollastonite, according to the thermal behavior of the materials. The initial endothermic peaks found at 70 and 80°C can be attributed to moisture evaporation from the paper, according to the results of differential scanning calorimetry of the acquired samples. The melting of the crystalline structures of cellulose derived from wheat and rice straw is represented by the second endothermic peaks, which are 202 and 205°C.

Differential Scanning Calorimetry (DSC)

In thermal analysis, the differential scanning calorimeter (DSC) is an essential instrument. It may be employed in a variety of sectors, including medicines, polymers, nanomaterials, and food. The data generated by these instruments is used to better understand amorphous and crystalline behavior, polymorph and eutectic transitions, curing and degree of cure, and a variety of other material characteristics utilized in product design, manufacturing, and testing.

The exothermic peaks on the DSC curve have broad temperature ranges that roughly match the weight loss ranges on the TGA curve. At 600°C, the paper containing wollastonite had a residual weight of roughly 3%, compared to paper without wollastonite, since wollastonite oxides do not break down at 600°C, but have a higher decomposition temperature. Due to the removal of considerable quantities of lignin, ash, and other minerals from the paper samples without wollastonite by delignification and hydrolysis, the residual weight of the paper samples without wollastonite reduced dramatically at 600°C. The findings show that wollastonite specimens with higher residual weight and thermal behavior have a higher decomposition temperature. The residues of wollastonite do not constitute an environmental danger since wollastonite is a natural substance formed when paper decomposes.

Biodegradability

The paper samples started to lose their original form after 10 weeks in the soil. The paper has degraded. Both paper samples were damaged when retrieving samples from the dirt. Figure 5 shows photographs of papers before and after decomposition during two months.

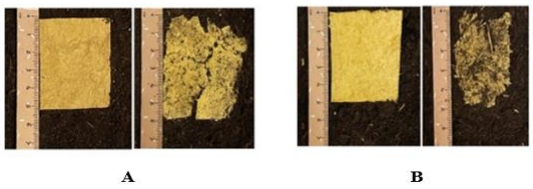

Fig. 5. Results of soil exposure before (A) and after (B) soil exposure.

The average weight loss from the initial dry weight of the straw-based material was 32% during 8 weeks of composting (Table 3).

After eight weeks of exposure to soil, more than 30% of the material based on 70% of the straw decomposes.

Due to moisture in the soil, biodegradation occurs at the edges of the samples, the paper loses its defined boundaries, fractures form, and industrial wrapping paper loses its original appearance. Naturally starch-coated paper degrades more quickly since starch is a suitable nutrition substrate for soil microbes [35]. The explanation for the decomposition of straw paper: it is well known that the soil contains a large number of bacteria and fungi that may degrade cellulose to monomers. Some Clostridia, such as Clostridium *thermocellum* [36], have been extensively studied for anaerobic cellulose breakdown [37], although they are already the subject of previous investigations.

Table 3. Weight loss of samples during 8-week composting

Sample	Average weight loss,%
Prepared wrapping paper from straw with a carton	32.1 ± 2.28
Industrial wrapping paper	29.8 ± 3.57

After eight weeks of exposure to soil, more than 30% of the material based on 70% of the straw decomposes. Due to moisture in the soil, biodegradation occurs at the edges of the samples, the paper loses its defined boundaries, fractures form, and industrial wrapping paper loses its original appearance. Naturally starch-coated paper degrades more quickly since starch is a suitable nutrition substrate for soil microbes [35]. The explanation for the decomposition of straw paper: it is well known that the soil contains a large number of bacteria and fungi that may degrade cellulose to monomers. Some Clostridia, such as Clostridium *thermocellum* [36], have been extensively studied for anaerobic cellulose breakdown [37], although they are already the subject of previous investigations.

The accelerated degradation of starch-coated paper is understandable: starch is a good nutrient medium for soil microorganisms [35]. The explanation that paper from straw has decomposed, it is known that there are a huge number of bacteria and fungi in the soil, so they can decompose cellulose to their monomers. Some Clostridia, for example, Clostridium *thermocellum* [36, 37], have been thoroughly investigated for the anaerobic decomposition of cellulose [37, 38], but these are already other studies.

A model was created based on the preceding findings: a Pareto chart. We investigated the complete dynamics of packaging paper's qualities by examining it. On the Pareto chart, the data affecting the quality of the packaging paper is analyzed (Figure 6). The length of the fibers has the most impact on papermaking, which is directly influenced by the speed and time of grinding, as well as the content of the paper pulp.

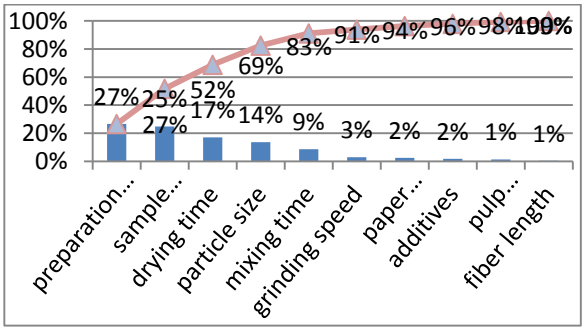

Fig. 6. Pareto chart for circumstances affecting paper quality.

The period of pulp boiling, the amount of cooked acid in the pulp, the cooking temperature, and the technological regime of grinding, casting, pressing, and drying, among other things, all influence each of these variables. The link between cellulose fibers, their composition, and the amount of basic and auxiliary ingredients injected all have a significant impact on the quality of paper (fillers, adhesives, and other substances) [39]. The process of delignification has a big influence on the qualities we've been talking about. A carbohydrate complex is present in the fractions of hemicellulose and low molecular weight cellulose, which increases the fibers' cohesive ability during boiling and their sealing ability in the wet state, i.e. the capacity to form links between neighboring fibers during swelling. Each of these factors is determined by the duration of pulp cooking, the content of boiling acid, the cooking temperature, the technological regime of grinding, casting, pressing, drying, etc. A great influence on the quality of paper is primarily exerted by the bond between cellulose fibers, the composition of the fibers, the amount of the introduced basic and auxiliary substances (fillers, adhesives, and other substances). The delignification method has a significant impact on the discussed properties. For the fractions of hemicellulose and low molecular weight cellulose, a carbohydrate complex is characteristic, which provides an increase in the cohesive ability of the fibers during boiling and the sealing ability in the wet state, i.e. bonds between adjacent fibers during swelling.

4. Conclusions

The best conditions for extracting cellulose from annual non-woody plants were chosen wheat and rice straw, and paper was made from the pulp, which not only had the required physical properties but was also biodegradable. FTIR spectroscopy was used to identify the functional groups of cellulose, and the structure of the resultant cellulose was established. TGA and DSC techniques were used to evaluate the thermal characteristics of the produced materials, which are equivalent to those of wood cellulose. Biodegradability is improved when packing paper is coated with starch, which is due to the nature of microorganisms that feed on starch and, as a result, cellulose decomposes to its monomers.

Wollastonite has been effectively used in the production of biodegradable packaging from natural biomass, such as agricultural and solid waste. The findings also show that wollastonite specimens with higher residual weight and thermal behavior have a higher decomposition temperature. The residues of wollastonite do not constitute an environmental danger since wollastonite is a natural substance formed when paper decomposes. Furthermore, the paper's overall strength has been enhanced. The paper becomes stiffer and more elastic. Additionally, after 10 weeks in the

soil, the paper samples began to lose their original shape. The quality of the paper has visibly deteriorated. When recovering samples from the soil, both paper samples were destroyed.

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Conflicts of interest

There are no conflicts to declare.

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