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The Technical and Economic Feasibility for the Production of Cellulose from Non-Wood – Agricultural Residues

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ABSTRACT

The paper is presented results of the technical and economical evaluation of the production of cellulose for the solving the problem of utilization of agricultural residues of lignocellulosic nature. Non-wood was used in this work for pulping. Treatment was proposed to carry out with the application of peracetic acid. Such method allows to obtain cellulose, lignin, furfural and xylose. Spent peracetic solution can be used to precipitate lignin and for regeneration of acetic acid and to recover furfural and xylose. A technological scheme for the processing of plant raw materials by this method has been developed. Economic efficiency was calculated. The sale of by-products can provide additional financial benefit. Such an approach in the processing of significant volumes of agricultural waste is effective from an economic point of view, as well as from an environmental point of view, as it allows to reduce the negative impact on the environment and to ensure its protection.

Keywords: non-wood, peracetic acid, cellulose, lignin, furfural, xylose.

INTRODUCTION

Environmental pollution as a result of the development of various branches of the chemical industry has reached a significant scale (Trus et al., 2020). A large negative impact on the environment is also associated with the development of agriculture, where, in addition to livestock waste, a large amount of plant waste is generated. Various innovative methods of improving the environment have been developed (Radovenchik et al., 2021). But the most effective is the prevention of the formation of toxic and dangerous compounds in technological processes, the development and implementation of environmentally safe technologies, the development of effective methods of disposal of by-products and waste.

Plant residues are a lignocellulosic biomass, which mainly consists of cellulose (40–60%), low-molecular weight polysaccharides (10–40%) and aromatic substances (15–30%), small quantities of extractive substances and minerals (Fahmy et al., 2017). Nowadays, the application of lignocellulosic residues from agriculture for obtaining different products, biofuels and chemicals is a modern direction of chemical technology (Gorobets and Karpenko 2017; Halysh et al., 2020). Chemical processing of straw and stalks of cereal and oil plants allows to prepare cellulosic fiber hat can be efficiently used for paper and cardboard production (Trembus et al., 2019). Today, this approach in processing of such non-wood materials is especially important for implementation in countries that have insufficient wood sources for cellulose production.

Conventional technologies of delignification with the application of chemicals based on sulfur compound are characterized by the emission of toxic substances. An important direction in development of delignification processes is the creation of resource-saving and ecologically friendly methods, e.g. organosolv treatment (Schulze et al., 2016; Shui et al., 2018).

The advantage of the method of obtaining cellulose by delignification with peracetic acid is that during the cooking process, acetyl groups are simultaneously split off as a result of hemicellulose hydrolysis. Thus, the formation of acetic acid takes pace (Kundu et al., 2021). There is no formation of methanol, as in the case of traditional methods of delignification. This facilitating solvent recovery, which is an important factor from an economic point of view. As a result of such delignification, it is possible to obtain lignin that does not contain sulfuric groups. At the same time, the hemicellulose fraction is transformed into marketable products (sugars and/or furfural). In the conditions of long-term delignification, furfural is the main byproduct of the hemicellulose hydrolysis reaction (Vila et al., 2003).

Technologies of organosolv treatment are described by different researchers but without taking into account technical and economical argumentations.

Therefore, the purpose of the research work was to evaluate the feasibility of agriculture residues utilization by peracetic treatment with the obtaining of cellulose and useful by-products.

MATERIALS AND METHODS

Evaluation of technical and economic feasibility were done based on the results of rapeseed straw peracetic cooking (Deykun et al., 2018). In laboratory conditions, after cellulose obtaining, the flask was cooled with tap water, the insoluble residue (mainly cellulose fiber) was filtered, and the spent solution was concentrated by evaporating 85% of its volume in a rotary evaporator. The concentrated residue was treated with 5 volumes of water from washing the cellulose product to precipitate the peracetic lignin. Precipitated lignin was separated from the filtrate by centrifugation for 10 min at 5500 rpm and washed with distilled water to the neutral value of the washing water, centrifuged again and dried at a temperature of 80 °C until the humidity reached 7–8%. Lignin yield was determined gravimetrically.

To perform the calculation of the technical and economic feasibility, we take into account the facilities and experience in the production of pulp products of PJSC "Zhydachiv Pulp and Paper Mill", Ukraine.

RESULTS AND DISCUSSION

The expediency of any technology connected with the of by-product processing and recovery of reagents of the solution (Amendola et al., 2012). The yield of the cellulosic product largely depends on the delignification parameters of plant biomass. The smaller the yield of the target product, the more components of the raw material are transferred to the cooking solution (Palamae et al., 2017). In the case of peracetic processing, the main components of the spent cooking solution are soluble low-molecular products of the

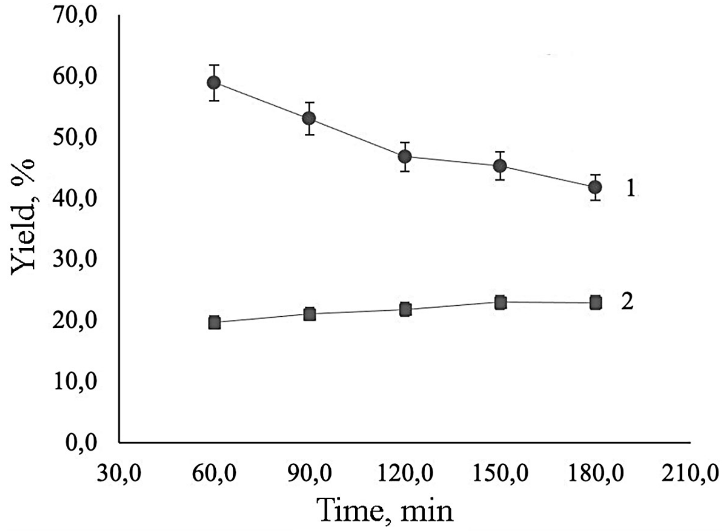

Figure 1. Dependence of cellulose and lignin yield on peracetic cooking time: 1 – cellulose; 2 - lignin

destruction of hemicelluloses and lignin, which is a promising material for obtaining a large number of valuable products (Dieste et al., 2016).

The results of investigation of the effect of time of peracetic processing of rapeseed straw on the yield of cellulose and lignin is shown in Figure 1. In the range of cooking from 30 to 180 min, the yield of cellulosic fiber decreased as the time prolonged. This happened due to delignification and partial degradation of polysaccharides components (Trembus et al., 2022). At the same time, the lignin content in spent solution is increased. As a result, the yield of precipitated lignin increased. Both materials can be used for further processing.

The technological scheme of agricultural waste processing by peracetic cooking is presented in Figure 2. The scheme also provides the recycling of spent solutions. According to the scheme, papered raw materials from the container (1) are load into the reactor (3), where the cooking solution from the cooking acid tank (2), containing glacial acetic acid and 30% hydrogen peroxide, is also dosed. The reaction mixture is heated to a temperature 95 °C. Time of the process is 2.0 hours. At the end of the cooking, the reaction mixture from the reactor (2) is fed to the screw press (4) to separate the spent solution from the cellulose product, after which the cellulose fiber is fed to the drum vacuum filter (5) for washing, and then for drying into the drying chamber (6). The spent solution is fed into the rotary evaporator (7), where 85% of the liquid phase evaporates. The resulting residue is sent to the mixer (8), where the washing water from the drum vacuum filter after pulp washing is added in a fivefold volume (5). As a result, the condensation of lignin occurs, which is then separated during centrifugation (9). The steam mixture from the rotary evaporator (7) enters the condenser (10), and then into the spent acetic acid tank (11), where acidic water obtained at the stage of precipitation and washing of lignin (9) is also supplied. The spent acetic acid solution mainly contains acetic acid, water, furfural, xylose, etc. substances in smaller quantities. It is fed to the distillation column (12), where furfural is separated, which is sent to the condenser (13) and for further processing. The solution from the lower part of the column is fed to membrane filtration to separate xylose and polysaccharide degradation products and to subsequent azeotropic distillation to obtain pure acetic acid.

Thus, a scheme has been developed that allows obtaining valuable primary cellulosic fiber, lignin, furfural and regenerating acetic acid for reuse in cooking processes.

The yield of the cellulosic product during 2 hours of cooking at a peracetic acid with concentration of 8.5% is 53.0%, then the calculation of the mass of straw for the to obtaining of 1 ton of cellulose with a moisture content of 10%, can be done according the formula:

$$m_{rm} = \frac{m_{cell \cdot K}}{Y_{cell}} \cdot 100 \quad (1)$$

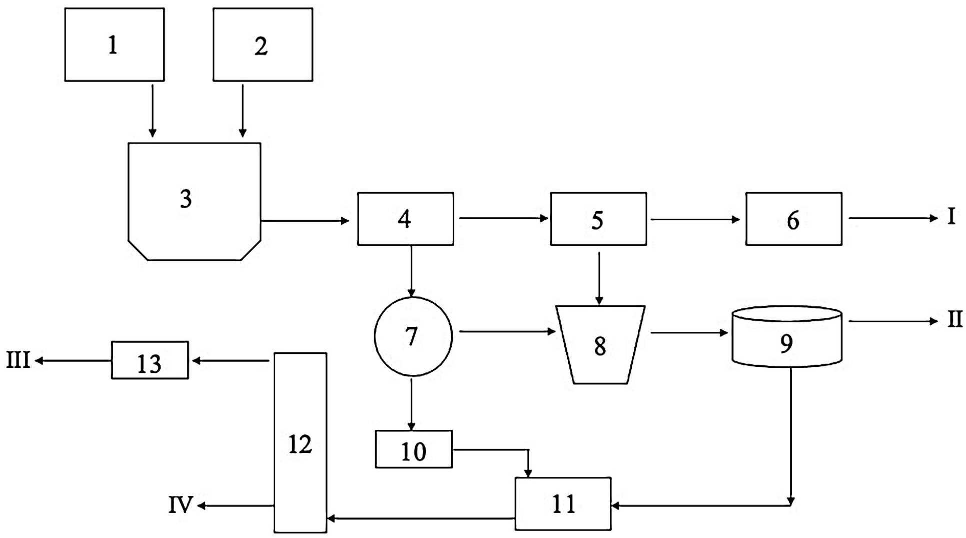

Figure 2. Basic technological scheme for agricultural waste processing: I – cellulose fiber for further processing; II – lignin-enriched solid residue for further processing; III – furfural; IV – acetic acid for membrane filtration and azeotropic distillation; 1 – container with raw material, 2 – tank of delignifying reagents with a dispenser, 3 – reactor, 4 – screw press, 5 – drum vacuum filter for washing cellulose product, 6 – drying chamber, 7 – rotary evaporator, 8 – mixer, 9 – centrifuge, 10 – condenser, 11 – spent acetic acid tank, 12 – distillation column

where: m_{cell} – mass of the oven dry cellulose, kg;
K – coefficient, which takes into account the dry substance content;
Y_{cell} – yield of the cellulose, %.

The mass of the oven dry raw material for the production of 1 ton of cellulose is:

$$m_{rm} = \frac{900 \cdot 0.9}{53.0} \cdot 100 = 1698.11 \text{ kg}$$

Then the mass of straw in an air-dry state with the humidity 12%, that must be taken:

$$\frac{1698.11}{0.88} = 1929.7 \text{ kg}$$

If we take into account that the yield of wood pulp by the sulfate method of aspen cooking is at the same level (Saltberg et al., 2009), then with a moisture content of the raw material of 12%, the required weight of wood to obtain 1 ton of cellulosic fiber with a moisture content of 8% is the same. In Ukraine, the cost of 1 ton of straw is 37.5 €, while the cost of 1 ton of wood is 58.5 €. The difference in cost is nearly 36%. If we take into account that the cost of bleached sulfate cellulose from hardwoods is 1175 € today, it is obvious that the cost of cellulosic fiber from non-wood will be lower. Taking into account the difference in the cost of raw materials, the estimated cost of 1 ton of primary cellulosic fiber from non-woods will be nearly 472 €.

The consumption of raw materials and their cost for the production of 1 ton of cellulosic fiber from non-wood is presented in the Table 1.

Taking into account that acetic acid can be completely regenerated in the process of processing spent solutions and returned to production, subsequent batches of cellulosic fiber production will be much cheaper. It is necessary to foresee the need to replenish possible losses of acetic acid with washing water in the amount of 2% of the total amount. Estimated cost of regeneration of cooking acid - 125 €. Then the cost of raw materials for the following batches of cellulose will be as shown in the Table 2.

The advantage of the peracetic cooking is that as a result of the process, lignin, furfural and xylose can also be obtained in addition to the cellulosic fiber. Depending on the cooking conditions, 390–400 kg of lignin, 115–121 kg of furfural and 50–59 kg of xylose can be obtained from 1 ton of pulp (Rahman et al., 2006; Rafiqul et al., 2014). The cost of these substances today is 6–10 €/kg, 45–50 €/kg and 10–15 €/kg, respectively.

The approximate cost of the products of peracetic cooking of non-wood is presented in the

Table 1. The consumption of raw materials and their cost for the production of 1 ton of cellulosic fiber from non-wood

Raw material/reagent	Consumption, kg	Price per unit, €	Total cost, €
Non-woods	1699	0.03	50.97
Acetic acid	8325.1	1.44	11988.14
Hydrogen peroxide	2752.4	0.66	1816.58
Water	5302.0	0.00060	3.22
Total			13858.92

Table 2. The consumption of raw materials and their cost for the production of 1 ton of cellulosic fiber from non-wood with regenerated acetic acid

Raw material/reagent	Consumption, kg	Price per unit, €	Total cost, €
Non-woods	1699	0.03	50.97
Acetic acid	8325.1	1.44	239.58
Hydrogen peroxide	2752.4	0.66	1816.58
Water	5302.0	0.00060	3.22
Regeneration costs	-	-	125.00
Total			2235.54

Table 3. Estimated cost of products of peracetic cooking of non-wood, €

Cellulose fiber per 1 ton	Lignin per 1 kg	Lignin per 1 kg	Lignin per 1 kg
625.00	3.25	5.00	2.00

Table 3. The sale of by-products can provide additional financial benefit.

The results of the calculation of the economic effect from the implementation of the method of utilization of the straw of cereal and oil crops indicate that, the necessary investment for the purchase of equipment is 70 000 €. Selling the cellulose product, lignin, furfural and xylose, obtained from 50 tons of straw, the investment payback ratio is 47%, and the investment payback period is less than 2 years.

CONCLUSIONS

It is shown that by peracetic cooking, non-wood can be effectively processed into cellulosic fiber to meet the needs of paper industries. The feasibility study confirmed the expediency of organizing the peracetic delignification process with the production of not only cellulose, but also the separation of delignification by-products such as lignin, furfural, and xylose, which are valuable substances. The reuse of regenerated acetic acid in the technological process ensures economical processing of raw materials. The calculation of the economic effect indicates the expediency of the application of peracetic cooking as an effective method of utilization of plant waste, which allows to obtain valuable substances of polysaccharide and aromatic nature.

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