

REALITIES AND PROSPECTS OF FUTURE COMPLEX PROCESSING OF PLANT RAW MATERIALS INTO BIOTHETANOL AND BY-PRODUCTS

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The use of plant biomass as a primary source of energy is currently unacceptable both from an economic and environmental point of view. The experience of a number of industries, in particular hydrolysis production, enables to solve the problem of profitability of organic biomass treatment by its deep complex processing with the resulting components whose cost exceeds the cost of organic raw materials as fuel. Currently, the main results of complex processing of organic raw materials are still energy-intensive products — bioethanol and hydrolyzed lignin, which energy characteristics are commensurate with fossil fuels. Bioethanol production from starch-containing, sugar-containing or lignocellulosic raw materials requires the use of different technological stages and, accordingly, the cost of bioethanol for each type of raw material is different. Compared to bioethanol produced from sugar and starch raw materials, bioethanol from manufactured lignocellulosic raw materials is more expensive. Bioethanol from lignocellulosic raw materials is more expensive compared to bioethanol obtained from sugar and starch raw materials. The most energy-intensive in the technology of bioethanol obtaining from lignocellulosic raw materials is the stage of pretreatment of raw materials for hydrolysis, because the process of preliminary preparation and hydrolysis with dilute acids occurs at high temperatures and pressures. During enzymatic hydrolysis, the process temperature is maintained for a long time (up to several days). To ensure deep integrated processing of plant raw materials, as well as to reduce overall costs, it was proposed to improve the technology and equipment, which allow increasing the degree of conversion of raw materials into basic and by-products.

Key words: bioethanol, biomass, preliminary preparation, hydrolysis, lignin, rotary pulsation apparatus.

1. Biomass as a renewable resource

The demand of the world economy for energy resources is growing every year. Thus, global oil reserves (as at 2015) amounted to 1657.4 billion barrels. Given that world oil consumption is estimated at 99 million barrels per day, there is a need for widespread use of renewable energy sources. Biomass is the largest renewable resource used in the world economy (more than 500 million tons per year) [1]. The most important sources of biomass are wood and crops.

The annual growth of biomass in the world is estimated at 146 billion tons in terms of

dry matter [2], which is energy equivalent to 80 billion tons of oil. The biomass potential in Ukraine is up to 20,2 million tons of oil equivalent [3]. These are straw, agricultural waste, waste from woodworking enterprises, etc.

Biomass is considered one of the key renewable resources of the future. Its amount reaches $2.423 \cdot 10^{12}$ t, or ≈ 550 Gt of carbon (C), of which $\approx 80\%$ is accounted for by plants, among which terrestrial plants are predominate [4].

Biomass is the most important source of energy for 3/4 of Earth's population living in developing countries. In developing countries, biomass provides an average of 38% of primary

energy (in some up to 90%). Biomass is likely to remain an important global source of energy in the 21st century. The most common sources of biomass are plants (wood, peat, straw). In the process of their use and processing, a large amount of renewable waste is generated every year. It is estimated that only 25% of waste is used efficiently. Developing countries, at the cost of biomass, can theoretically cover 15% of energy needs, industrial — 4% [5].

2. The state of bioethanol production in Ukraine and the types of raw materials available in Ukraine

In 2018, there were about 13 bioethanol producers in Ukraine. Six of them are quite successful — Zarubinsky, Gaisinsky distilleries, private plants Ecoenergy and Fazor, as well as Uzinsky and Gnidavsky sugar factories. In 2017, they produced 80,000 tons of bioethanol, which went to the production of alternative fuels [6].

Currently, industrial plants in bioethanol production employ mainly two types of primary raw materials, namely starch from cereals and juice or molasses from sugar crops (first generation bioethanol) [7].

Energy crops for ethanol production in the European climate zone, which includes Ukraine, are plants with high sugar and starch content — cereals, potatoes, sugar beets, corn for grain [8].

Corn has the highest yield of bioethanol per product unit — an average of 40 liters per 1 centner of grain (with dry grinding). The result of corn complex processing (wet grinding) is starch, gluten, glucose-fructose syrups, lysine, food alcohol, or bioethanol, citric and lactic acids, protein-mineral vitamin concentrates [9].

Another type of raw material that can be used for bioethanol production in Ukraine is sugar beet. This culture provides the highest yield of ethanol from 1 ha, does not require additional costs for pre-treatment of raw materials, allows reducing greenhouse gas emissions [10]. By-products of sugar production are beet pulp, which is used as fodder for agricultural livestock, molasses, and filtration sludge [11].

Studies on the use of energy crops as raw materials for bioethanol production are energy willow, miscanthus, sugar sorghum [12].

The optimal raw material for bioethanol production in Ukraine is molasses — a by-

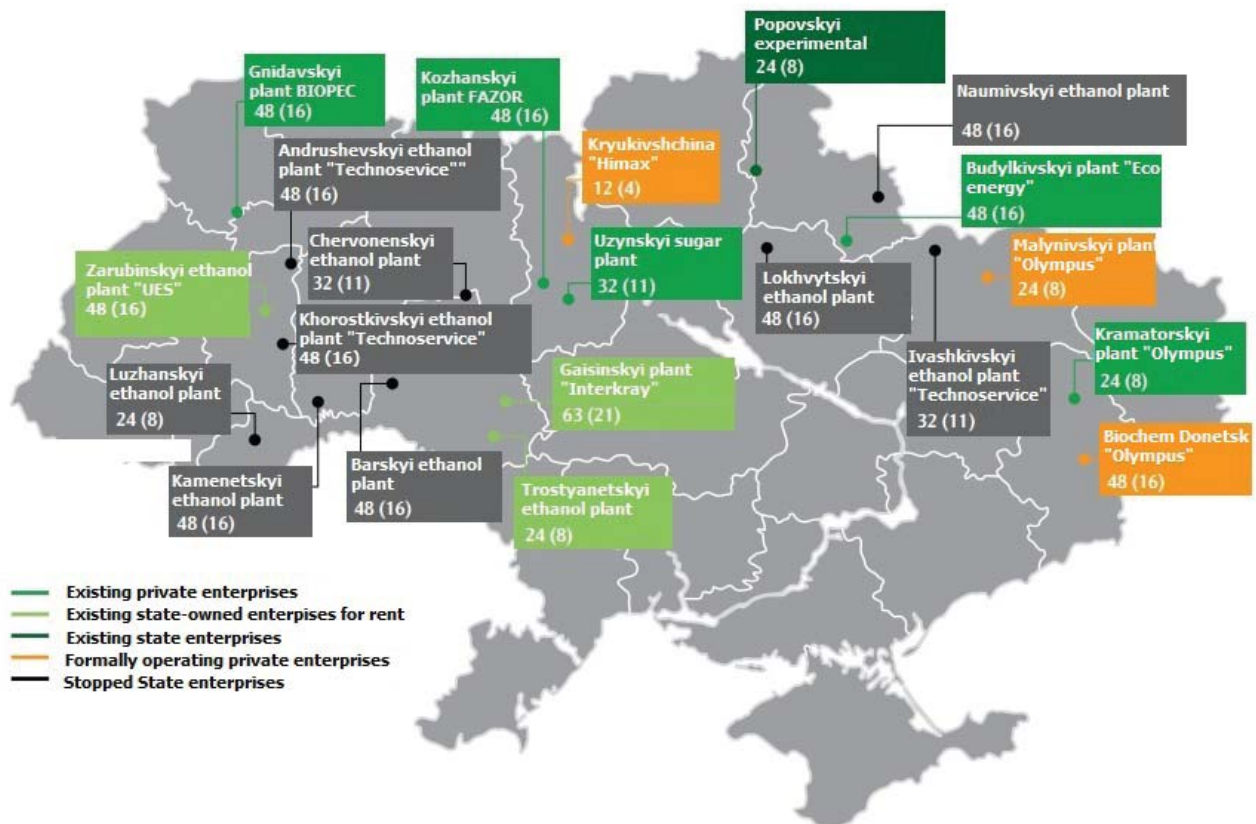


Fig. 1. Ukrainian enterprises of various forms of ownership that produce bioethanol (for 2018) [6]

product of sugar production. According to the authors of [13], the total energy consumption for one ton of bioethanol from molasses is 9 459 MJ, from corn — 56 565 MJ. The difference in production energy consumption is due to the presence of the stage of the operation of boiling raw materials and saccharification of starch by malt enzymes or enzyme preparations. The process of molasses preparation for fermentation consists only of homogenization of antiseptic molasses and dilution with water (spreading).

The main disadvantage of molasses as a raw material for bioethanol production is its limited amount. According to statistics, in 2019 the sugar factories of Ukraine received 832581.6 tons of sugar, 256067.6 tons of molasses, and 1297780 tons of pulp. That is, from 256067 tons it is possible to get 77896 tons of bioethanol, or $(77896 \text{ tons} \cdot 0.64 = 49853.44 \text{ tons of oil equivalent})$ [14].

However, molasses are also used as a raw material for the production of food acids, baking, and feed yeast, as an additive to farm animal feed, as well as a binder in the lumping of fine coal [15].

The simplified block diagram of complex processing of grain raw materials for bioethanol obtaining with designation of by-products are shown in Fig. 2.

The comparative characteristics of different types of biomass as raw materials for bioethanol production are shown in Table 1.

3. Economic efficiency of different methods for bioethanol obtaining

It should be noted that the use of plant biomass as a primary energy source is currently unacceptable from both an economic and environmental point of view. The experience of a number of industries, in particular hydrolysis, allows solving the problem of profitability of organic biomass processing by its deep complex processing to obtain components whose cost exceeds the cost of organic raw materials as fuel.

The main results of the complex processing of organic raw materials are increasingly energy-containing products — bioethanol and hydrolyzed lignin, which energy characteristics are commensurate with fossil fuels.

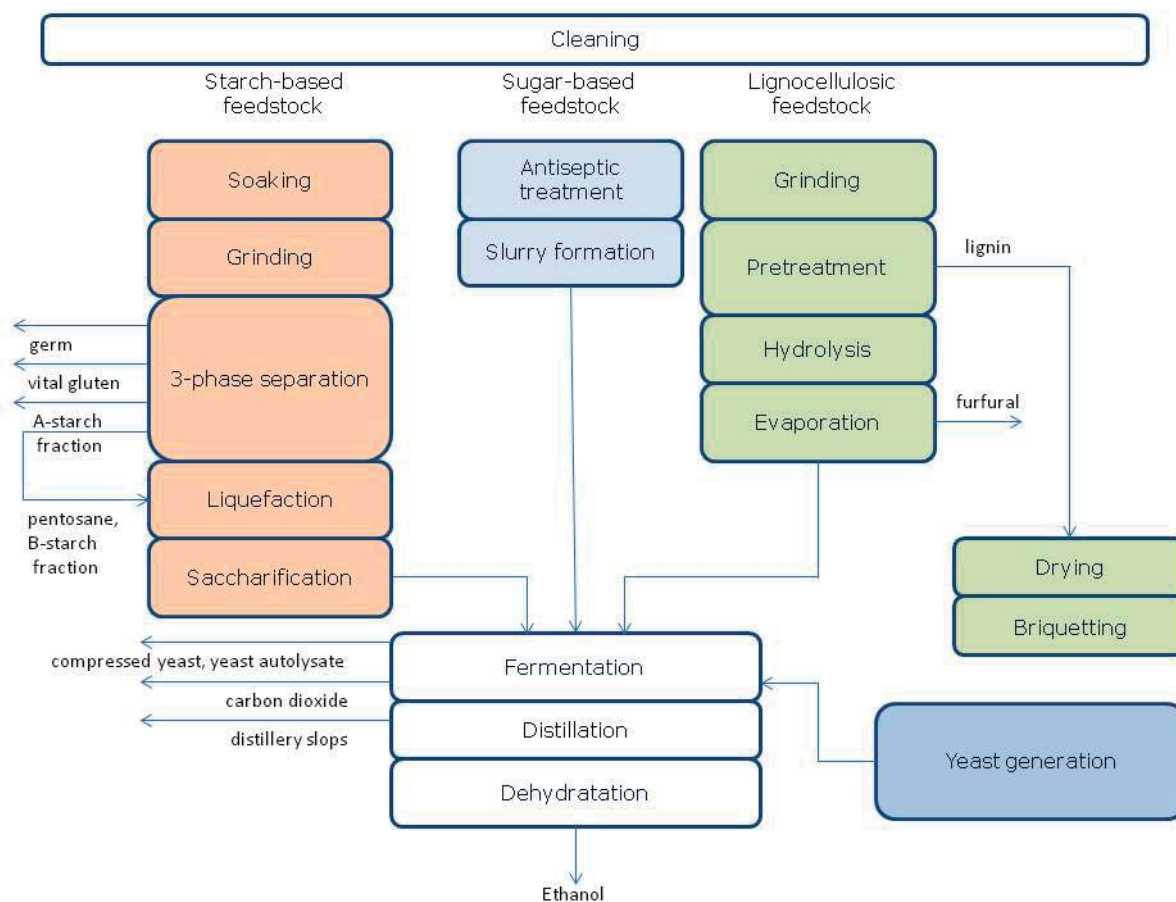


Fig. 2. Block chart of complex processing of grain raw materials and molasses for bioethanol obtaining with by-products designation

Table 1. Technological evaluation of raw materials for bioethanol production in Ukraine [16]

Type of raw material	Yield of bioethanol, th/tons of raw material	The required sowing area for the production of one ton of bioethanol, ha
Spiked cereals	0.3	1.24
Beet	0.08	0.42
<i>Sorghum vulgare</i> var. <i>saccharatum</i> , <i>Sorghum saccharatum</i>	0.09	0.14
Molasses	0.24	–
Wheat straw	up to 0.1*	–

Note: enzymatic hydrolysis with pretreatment by purified KOH [17].

The main direction of improving the process of complex processing of organic biomass, in particular plant raw materials, is the introduction of technologies and equipment that increase the degree of conversion of raw materials into basic and by-products without a significant increase in energy consumption.

The main factor that determines the efficiency of bioethanol production is energy consumption per unit of output.

Table 2 shows the average energy consumption of the plant for 1000 liters of bioethanol production. These indicators are typical but may vary depending on specific local conditions (according to VOGELBUSH Biocommodities) [18]. Steam consumption and supplied power vary depending on whether the drying of the post-alcoholic bard (wine concentration) takes place or not.

From the data given in Table 2 it can be concluded that the production of bioethanol from molasses is 44% less energy consuming compared to the production of bioethanol from wheat. The additional operation of drying the post-alcoholic bard, offered by the manufacturer, adds about 60% to energy

consumption (for molasses) and more than twice as much for wheat.

As an example of efficiency of deep grain processing, the material balance of typical plants offered by the VOGELBUSH group of companies is given (Table 3).

There are a number of studies on the cost-effectiveness of processing for various types of plant raw materials in order to obtain sugar with subsequent production of ethanol and by-products [20–22], in particular in [22] the fixed capital costs (buildings, equipment and plant construction costs, and production costs (including materials, labor, utilities, maintenance fees, and capital depreciation) (Fig. 3).

The figure shows that when investing in fixed capital, pulp processing has higher fixed costs than that of first-generation raw materials.

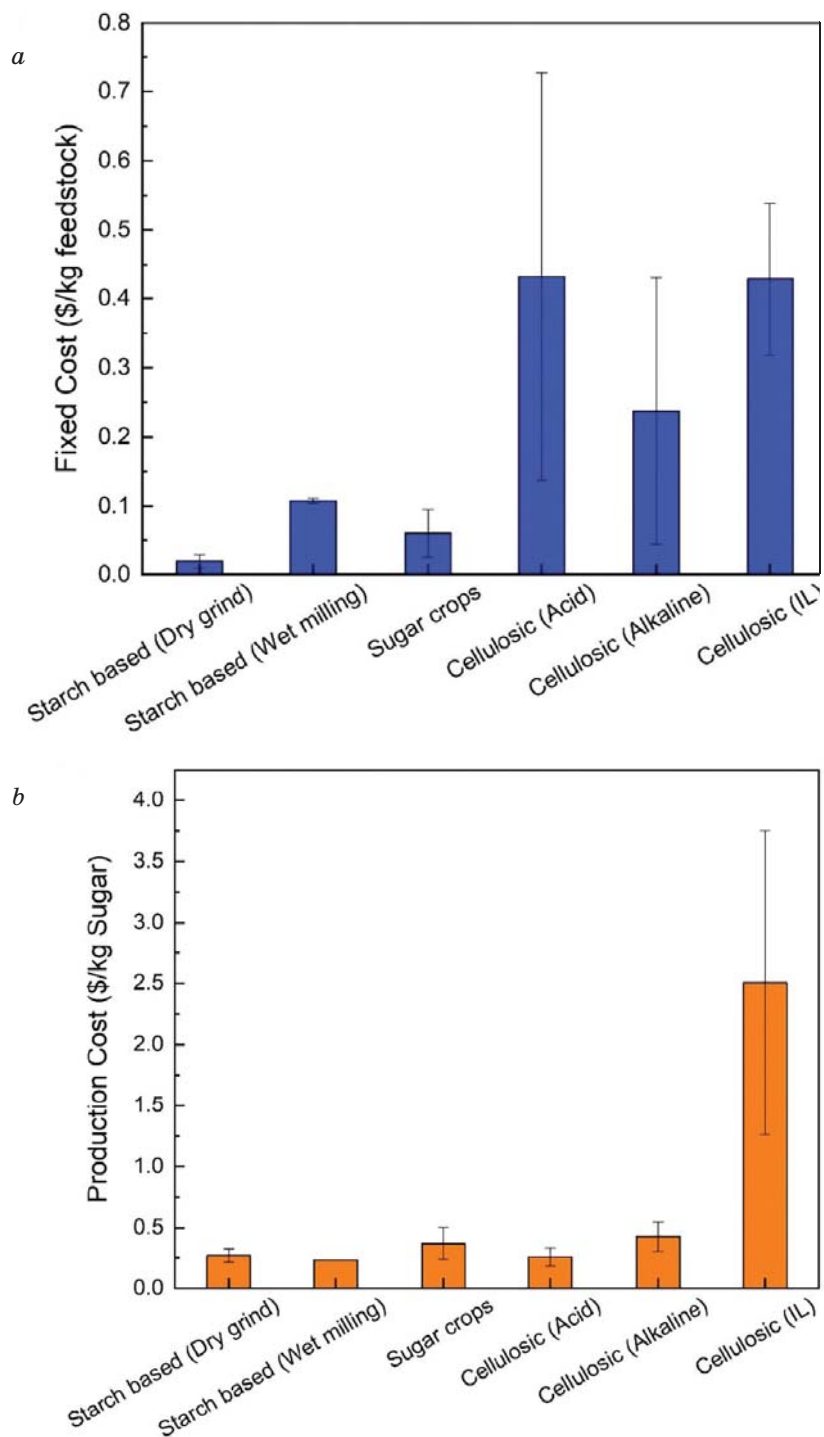
The acid-based pretreatment has the highest fixed cost because of the extra costs required to guard against equipment corrosion. The ionic liquid and organosolv pretreatments have the second highest fixed costs because of the additional equipment needed for recycling

Table 2. Average energy consumption of the entire plant for 1000 liters bioethanol production (according to VOGELBUSH Biocommodities)

Raw material	Part of starch (sugar*)	Quantity, kg	Steam, kg		Power, KW/h	
			Drying of post-alcoholic bard or vinassa concentration			
			no	yes	no	yes
Wheat	62	2420	1400	3150	115	260
Cereals	65	2285	1250	2750	110	220
Molasses	52*	3210	1700	3000	65	105
Cane juice <i>Sorghum vulgare</i> var. <i>saccharatum</i> , <i>Sorghum saccharatum</i>	18*	8640	1200	2550	60	90

Table 3. Added value of typical plants (according to the VOGELBUSH group of companies) [19]

Type of enterprise	Wheat, thousand tons/year	Added value, million euros/year
A simple plant for grain processing into bioethanol	500	47.7
Deep processing plant grains	500	77.1
Plant for deep grain processing and bioethanol production	1000	123,3

Fig. 3. Costs of sugar production from different types of feedstocks: *a* — fixed capital cost; *b* — production cost [22]

and facilities for storing the solvents. On the other hand, the least expensive sugar production operation used sugar crops because it required a relatively simple process for juice extraction.

Fixed costs for the wet grinding process are higher than for the dry grinding process due to operations related to the fractionation of corn grain.

Equipment used for raw material processing, pretreatment, and lignin separation is a major factor in increasing fixed capital investment for pulp raw materials.

As it is shown in Fig. 3, *b*, pretreatment of raw materials before hydrolysis using acid has the lowest cost of sugar production, because it has high efficiency and productivity of sugar. Lower sugar alkaline pretreatment productivity leads to higher sugar production costs.

The most promising lignocellulosic raw materials for complex processing are agricultural waste. Thus, in 2017, primary crop waste (straw and stems) amounted to 15039 thousand tons of oil equivalent, or 57.7% of the total biomass potential of agricultural enterprises of Ukraine, wood biomass — 16.3%, energy crops — 14.5% [23].

Fig. 2 shows that the common stages of the technological process of bioethanol obtaining, for these types of raw materials are the generation of yeast, fermentation, distillation, and dehydration of ethanol. The differences of the previous stages are determined by the type of raw material, the method for obtaining from it a sugar solution for fermentation and byproducts obtaining from it that are not used in ethanol production.

The most energy-intensive is the method of bioethanol obtaining from lignocellulosic raw materials, which consists of a stage of pretreatment for hydrolysis and direct

hydrolysis. The main energy consumption is to maintain high temperatures and pressures during pretreatment and hydrolysis with dilute acids and to maintain a certain temperature for a significant time (up to several days) during enzymatic hydrolysis.

The main purpose of pretreatment of lignocellulosic raw materials for hydrolysis is the removal of lignin, which prevents the access of cellulolytic enzymes to cellulose fibers. Commercial pretreatment technologies are shown in Table 4 [24].

Cost distribution for the general lignocellulosic enterprise equipment is given in [24]. Pretreatment and steam production are the most cost-effective areas.

One of the ways to increase the efficiency of complex processing of lignocellulosic raw materials is the application of equipment that enables to increase the degree of raw materials conversion into basic and by-products. An example of the equipment that significantly intensifies heat and mass transfer in liquid multicomponent media are rotary pulsation devices, which are effective devices in technologies related to mixing, homogenization, dispersion, etc. [25]. In particular, there are a number of studies on the use of rotary pulsation devices in the technology of making wort from starch-containing raw materials [26, 27].

The research results on the use of rotary pulsation apparatus for delignification of wheat straw in the process of its preliminary preparation for hydrolysis are given in [28]. The hardware-technological scheme of the pretreatment stage for hydrolysis using a rotary pulsation apparatus and alkali as an agent for removing lignin is shown in Fig. 5. The receiving tank 1 and the rotary pulsation apparatus 2 are filled with an alkaline solution. After the engine start, the pre-crushed chips

Table 4. Commercial technologies of of lignocellulosic raw materials prearrangement for hydrolysis

Type of process	Manufacturer	Characteristic
Steam explosion	Beta Renewables	Low xylose yield A large amount of enzyme preparation
One-stage treatment with dilute acid	Abengoa	High yield of xylose medium amount of enzyme preparation
Two-stage treatment with diluted acid	Poet–DSM	High yield of xylose Small amount of enzyme preparation
Ammonia and steam	Dupont	Requires a large amount of enzyme preparation

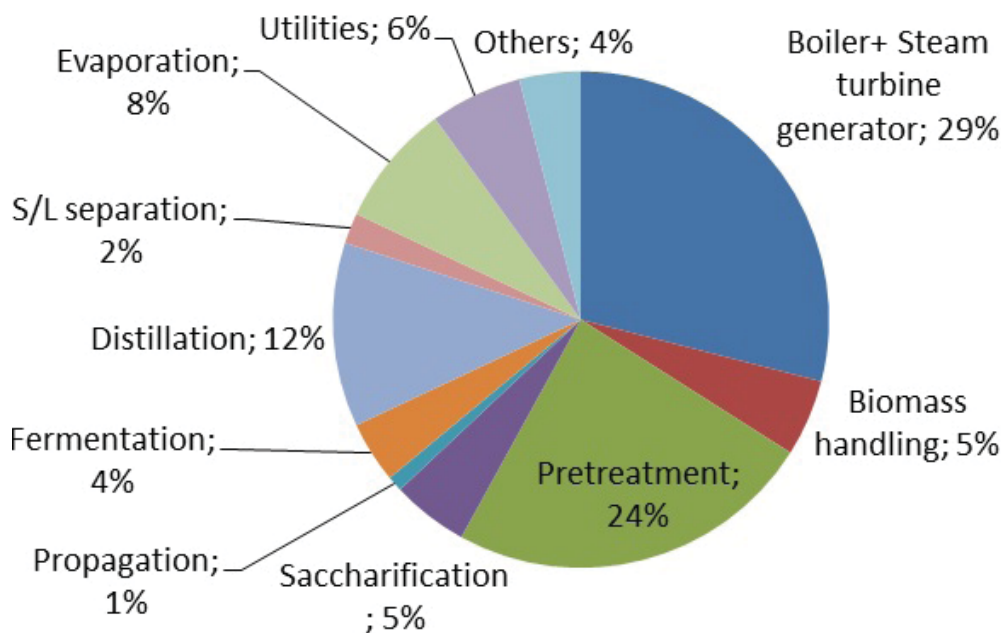


Fig. 4. Cost distribution for equipment of the enterprise for the bioethanol production from cellulose-containing raw materials [24]

of annual plant raw materials are added into the solution moving in the reverse flow. The resulting dispersion passes through a rotary-pulsation apparatus. Processing is carried out in the recirculation mode at certain temperatures (up to 100 °C) and pressure (up to 0.1 MPa). The resulting dispersion is fed to filter 3 (optionally diluted with water), in which the filtrate containing lignin and the solid cellulose residue, which is fed to the stage of hydrolysis, is separated.

The filtrate from the filter is fed to the intermediate tank 4 and subjected to chemical treatment with sulfuric acid up to pH = 2, after which it is fed to filter 5. The precipitated lignin remains on the filter and is fed to the drying stage. After filter 5 the filtrate is fed to the stage of hydrolysis.

If bioethanol is considered as an item, it has a stable high demand and turnover, which has a profitability of 15–30%. The production building is quite compact 30×21×24 meters. Of course, by building a complex for bioethanol production, the company has the opportunity to modernize processing and launch the production of baker's yeast, furfural or liquefied carbon dioxide, which are sold as separate products. Among the options for the future, the construction projects are considered for biogas production facilities through the methanation of bard and pulp with the production of green electricity, which

is implemented with a factor of 2.2–2.5. Every day, the plant, which processes 200 tons of molasses, produces 50 tons of bioethanol, 40 tons of carbon dioxide [29].

Lignin and bioethanol are energy-intensive products of plant biomass processing. The calorific value of dry lignin is 5500–6500 kcal/kg and is comparable to the calorific value of equivalent fuel (7000 kcal/kg). The caloric content of the product with a moisture content of 18–25% is 4400–4800 kcal/kg and the value of the product with a moisture content of more than 65% is 1500–1650 kcal/kg, the caloric content of ethanol is respectively 6405 kcal/kg.

27 thousand tons of fuel equivalent per year could be produced from 43 thousand tons of absolutely dry lignin with a moisture content of 60%.

According to the world leader in lignin production Borregaard, 400 kg of cellulose, 50 kg of ethanol, 3 kg of vanillin, and 400 kg of hydrolyzed lignin are obtained from 1000 kg of wood after autoclaving, decolorization and drying.

According to various sources, the amount of lignin in Ukraine (waste from the yeast, feed, pulp, and paper industry) is from 5 to 15 million tons.

It is known that deep complex processing of plant raw materials can significantly increase the profitability of the relevant industries by obtaining additional products with high added

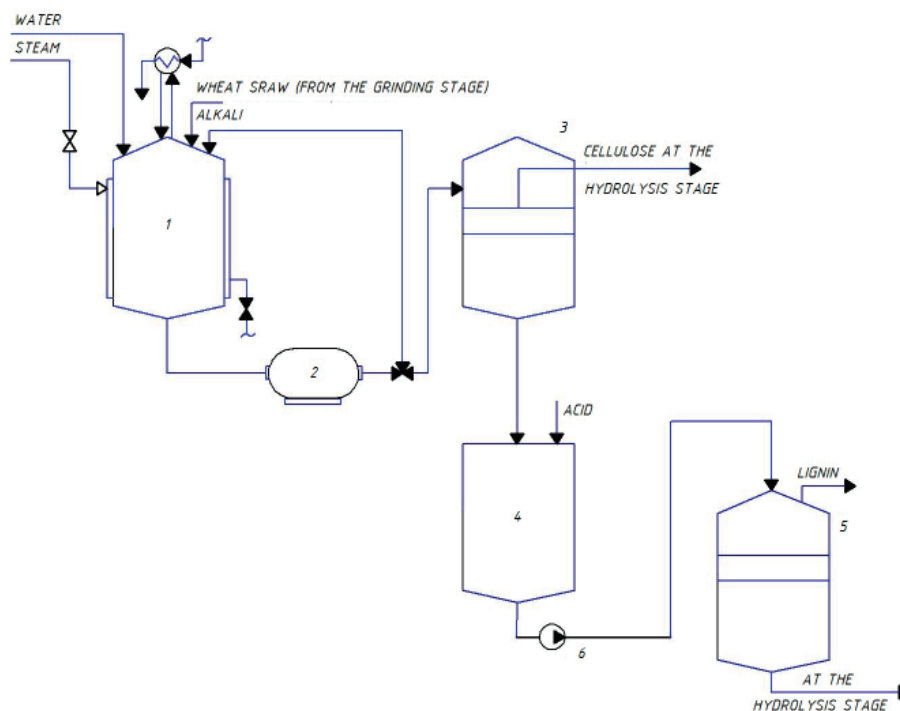


Fig. 5. Hardware-technological configuration of the pretreatment stage for hydrolysis using a rotary-pulsation apparatus:

1 — receiving capacity; 2 — rotary pulsation apparatus; 3, 5 — filter; 4 — intermediate capacity; 6 — pump

value, thus reducing the cost of basic products and increasing the profitability of production as a whole. So, the substances obtained in the process of chemical destruction of organic raw materials in hydrolysis or yeast production are in great demand in the pharmaceutical, chemical, cosmetic industry, construction, and so on [5]. Much attention is paid to the integrated processing of organic raw materials in industrialized countries.

The centers for integrated processing of organic raw materials operate in Austria, France, Denmark, Germany, Canada, the Netherlands.

Thus, the Austrian company “Lenzing AG” produces furfural, acetic acid, sodium sulfite, potassium, lignin. At the same time, thermal and electrical energy is produced on the basis of lignin as an energy carrier.

In Canada, “Ensyn” recycles crop and wood waste, produces biodiesel, charcoal, binders, “green gasoline” (bioethanol), diesel, and aviation fuel (rapid pyrolysis is used). Biodiesel is used to produce heat and electricity, motor fuels, and a number of chemicals.

GEA Wiegand GMBH from Germany, producing bioethanol, additionally produces gluten flour, starches, dextrose, fructose syrup, and other high-value products.

Lignins, as products of complex processing of plant raw materials, have been used as dispersants, emulsifiers, binders and adhesives, sorbents for medical and technological purposes, sorbents for wastewater treatment [30, 31].

In the pharmaceutical field, lignin is considered as a promising material for hydrogels production, because lignin has antioxidant and antibacterial properties [32], it can be used as a material for 3D printers for the manufacture of dressings for wounds. The components of lignin decomposition are considered as antiviral drugs and antioxidants [33].

However, commercially lignins continue to be used as biofuels (fuel briquettes, pellets, including in a mixture with sawdust, coal and peat dust, fuel gas production, including the production of electricity in gas-piston gas generators, boiler fuel, etc.).

The most common type of use of hydrolyzed lignin with a humidity of 60–65% is its combustion to produce heat.

Modern operating enterprises for lignocellulosic raw materials processing use lignin obtained during processing as fuel for steam production at the enterprise.

The use of lignin and fuel quality can cover up to 73% of the thermal energy demand for bioethanol production in an alkaline way [34].

Bioethanol obtaining from starch-containing, sugar-containing or lignocellulosic raw materials requires the use of different technological stages so the cost of bioethanol for each type of raw material is different accordingly. Compared to bioethanol with sugar-containing and starch-containing raw materials, bioethanol from lignocellulosic-containing raw materials is more expensive. The most energy-consuming is the stage of preliminary preparation of raw materials for hydrolysis in the technology of bioethanol obtaining from lignocellulosic raw materials. Increasing the profitability of bioethanol

production from lignocellulosic raw materials is possible in two main ways, namely by its deep complex processing to obtain components whose cost exceeds the last one of organic raw materials as fuel and technology and equipment improvement enabling to increase the conversion of raw materials into basic and by-product materials.

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РЕАЛІЇ СЬОГОДЕННЯ ТА ПЕРСПЕКТИВИ КОМПЛЕКСНОЇ ПЕРЕРОБКИ РОСЛИННОЇ СИРОВИНИ НА БІОЕТАНОЛ ТА ПОБІЧНІ ПРОДУКТИ

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Використання рослинної біомаси як первинного джерела енергії на сьогодні є неприйнятним ні з економічного, ні з екологічного погляду. Досвід низки виробництв, зокрема гідролізного, дає змогу вирішити проблему рентабельності оброблення органічної біомаси шляхом глибокої її комплексної переробки з отриманням складових, вартість яких перевищує вартість вихідної органічної сировини як палива. Наразі головними результатами комплексної переробки органічної сировини залишаються енерговмісні продукти — біоетанол та гідролізний лігнін, які мають енергетичні характеристики, співставні з викопними паливами. Отримання біоетанолу з крохмалевмісної, цукровмісної або лігноцелюзовмісної сировини потребує застосування різних технологічних стадій і, відповідно, собівартість біоетанолу для кожного виду сировини є різною. Порівняно з біоетанолом і цукровмісною та крохмалевмісною сировиною біоетанол з лігноцелюзовмісної сировини є дорожчим. Найенерговитратнішою в технології отримання біоетанолу з лігноцелюзовмісної сировини є стадія попередньої підготовки сировини до гідролізу, оскільки цей процес і гідроліз розведеними кислотами відбувається за високих температури та тиску. Під час ензиматичного гідролізу температура процесу підтримується протягом тривалого часу (до кількох діб). Для забезпечення глибокої комплексної переробки рослинної сировини, а також з метою зменшення загальних витрат запропоновано вдосконалення технології та обладнання, що уможливить збільшення ступеня конверсії вихідної сировини в основні та побічні продукти.

Ключові слова: біоетанол, біомаса, попередня підготовка, гідроліз, лігнін, роторно-пульсаційний апарат.

РЕАЛИИ НАСТОЯЩЕГО И ПЕРСПЕКТИВЫ КОМПЛЕКСНОЙ ПЕРЕРАБОТКИ РАСТИТЕЛЬНОГО СЫРЬЯ В БИОЭТАНОЛ И ПОБОЧНЫЕ ПРОДУКТЫ

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На сегодняшний день использование растительной биомассы как первичного источника энергии является неприемлемым ни с экономической, ни с экологической точки зрения. Опыт ряда производств, в частности гидролизного, позволяет решить проблему рентабельности обработки органической биомассы путем глубокой ее комплексной переработки с получением составляющих, стоимость которых превышает стоимость исходного органического сырья как топлива. Ныне главными результатами комплексной переработки органического сырья остаются энергосодержащие продукты — биоэтанол и гидролизный лигнин, имеющие энергетические характеристики, соизмеримые с ископаемыми топливами.

Получение биоэтанола из крахмалсодержащего, сахаросодержащего или лигноцеллюлозосодержащего сырья требует применения различных технологических стадий обработки и, соответственно, себестоимость биоэтанола для каждого вида сырья различна. По сравнению с биоэтанолом с сахаросодержащего и крахмалсодержащего сырья стоимость биоэтанола из лигноцеллюлозного сырья является более высокой. Наиболее энергозатратной в технологии получения биоэтанола из лигноцеллюлозосодержащего сырья является стадия предварительной подготовки сырья к гидролизу, поскольку этот процесс и гидролиз разбавленными кислотами происходит при высоких температуре и давлении. Во время энзиматического гидролиза температура процесса поддерживается в течение значительного времени (до нескольких суток). Для обеспечения глубокой комплексной переработки растительного сырья, а также с целью уменьшения общих затрат предложено совершенствование технологии и оборудование, позволяющее увеличить степень конверсии исходного сырья в основные и побочные продукты.

Ключевые слова: биоэтанол, биомасса, предварительная подготовка, гидролиз, лигнин, роторно-пульсационный апарат.