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LIGNOCELLULOSIC BIOMASS AS A FEEDSTOCK FOR THE CELLULOSE ETHANOL (2G) PRODUCTION

BIOMASA LIGNOCELULOZOWA JAKO SUBSTRAT DO PRODUKCJI ETANOLU CELULOZOWEGO (2G)

***Summary**: In the paper, the possibilities of utilizing the lignocellulosic biomass in the second generation bioethanol (2G) production were presented. The most important groups of lignocellulosic raw materials were characterized. The composition and structure of biomass and the methods for its conversion to ethanol were described. Moreover, the conceptions of utilizing the lignocellulosic biomass not only as the renewable energy source for production of biofuels but also of other products with the value added within the frames of integrated technological processes in biorefineries, with the consideration of the estimated costs of cellulose ethanol production were presented.*

***Keywords**: lignocellulosic biomass, biofuels, cellulose ethanol, the second generation (2G) bioethanol, biorefinery*

***Streszczenie**: W pracy przedstawiono możliwości wykorzystania biomasy lignocelulozowej do produkcji bioetanolu drugiej generacji (2G). Scharakteryzowano najważniejsze grupy surowców lignocelulozowych. Opisano skład i budowę biomasy oraz metody jej konwersji do bioetanolu. Ponadto zaprezentowano koncepcje wykorzystania biomasy lignocelulozowej nie tylko jako odnawialnego źródła do produkcji biopaliw, ale również innych produktów o wartości dodanej w ramach zintegrowanych procesów technologicznych w biorafineriach, z uwzględnieniem szacunkowych kosztów wytworzenia etanolu celulozowego.*

***Słowa kluczowe**: biomasa lignocelulozowa, biopaliwa, etanol celulozowy, bioetanol drugiej generacji (2G), biorafineria*

Introduction

The demand on energy has been systematically increasing for many years. It results, first of all, from the increasing number of population as well as due to the intensive development of industry. At present, the energetic needs are mainly satisfied by the traditional fossil fuels, however, their incineration is not neutral for the environment. The greenhouse gases (GHG) as emitted during combustion, constitute the greatest problem [1]. At the present moment, about 20% of the world energy is used on the electric energy whereas 80% are spent for the fuels [2]. In 2015, the transport sector itself was responsible for 19% of the world final demand on energy; the majority of it derived from fossil fuels [3].

One of the methods of climate changes' mitigation, with the simultaneous ensuring the energetic safety is production of alternative transport fuels [4]. The most important are biodiesel and bioethanol which is a substitute and bio-component for petrol. Bioethanol is produced first of all from the saccharose or starch-abundant raw materials [5]; however, their utilization stays in collision with the production of food and feeds [6]. They are the so-called first generation fuels, the participation of which in renewable energy in transport up to 2020 has been limited up to 7% in the Directive of the European Parliament and of the Council 2015/1513 of September 9, 2015. The mentioned Directive contains also the records concerning the necessity of supporting the studies in respect of advanced biofuels, including bioethanol of the second generation (2G).

The biofuels, produced from the lignocellulosic materials generate a low net emission of GHG and by this, decrease a negative effect on the environment, counteracting the unfavourable climate changes [7]. Production of advanced biofuels decreases the dependence on the fossil fuels, especially of crude oil import; it contributes also to the minimization of negative consequences for natural resources and food safety, supply and quality of water and soil on the local, regional and global levels [8].

In 2016, the European Commission submitted the legislative proposal, changing the Directive on the renewable energy sources and laying down the policy for the period after 2020. According to its assumptions, the participation of traditional biofuels should be further gradually decreased from 7% to 3.8% in 2030; the advanced biofuels should increase its share to 1.5% in transport fuels in 2021 and then, every year at least to 6.8% until 2030. According to the report of the International Renewable Energy Agency [9], 22% of transport fuels in 2050 will come from liquid biofuels and biogas.

Lignocellulosic raw materials

Bioethanol is produced from biomass, containing polysaccharides or carbohydrates which may be transformed into fermentable sugars. They are fist of all, sugar crops and by--products coming from sugar refining, starch-containing plants; it may be also lignocellulosic biomass [10] which is not only the renewable but also abundant source [Fig. 1). It is a sustainable alternative for petroleum-derivate fuels, it is universally available and reveals a smaller competition with production of food and feeds as compared to the substrates used for production of the first generation biofuels [11].

The complex structure (Fig. 2) is one of the main technical and economic barriers to utilization of lignocellulosic biomass as a raw material in production of biofuels [13]. The lignocellulosic biomass contains first of all polysaccharides, including cellulose (20–50%) and hemicellulose (15–35%), aromatic polymer – lignin (5–30%) [14, 15] and extracts and ashes [13]. Cellulose is the most popular natural polymer with a linear chain composed of glucose molecules. It consists of 100–1000 units, connected with β-1,4- glycoside bonds [16]. The cellulose chains constitute the so-called fibrils. The cellulose fibers, linked with the hydrogen bonds [17] are found in the lignocellulose matrix what makes that they are very resistant to enzymatic hydrolysis [18]. Hemicellulose creates the branched chains, composed of xylose molecules (usually C5 sugars). The degree of polymerisation of hemicelluloses is lower in comparison to cellulose and is found within the range of 100–200 units [19]. They act as physical barrier, limiting the availability for the enzymes [18]. Lignin is a heteropolymer composed of phenol alcohols' derivates, including p-coumaryl, coniferyl and sinapyl alcohols [20]. Lignin blocks the access of enzymes to cellulose; it may also irreversibly adsorb cellulases [18]. It is resistant to chemical and biological degradation. The presence of lignin in lignocellulosic substrates makes the fermentation process difficult in a considerable degree [21].

Fig. 1. The main groups of cellulosic feedstock [12]

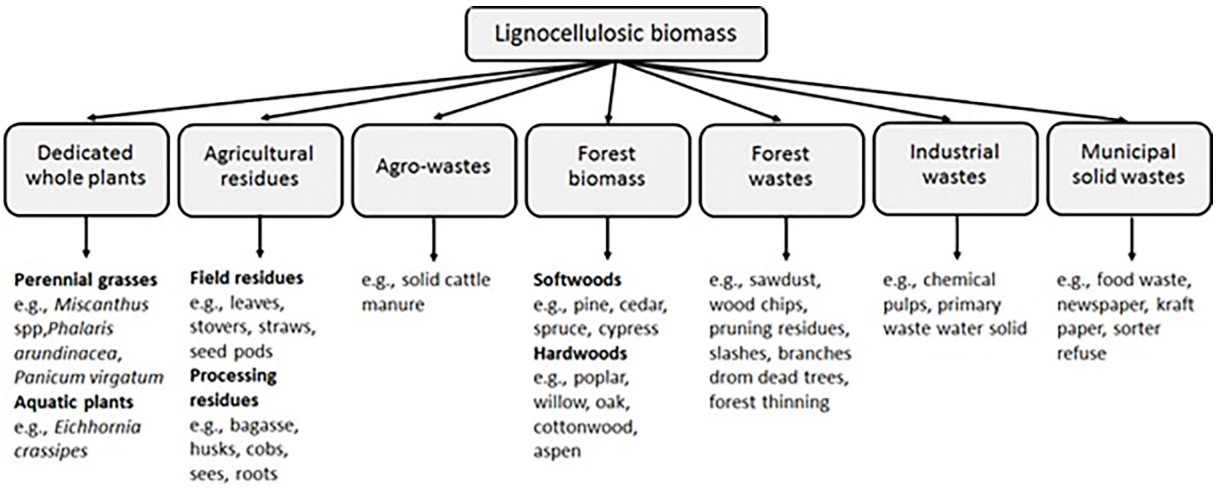
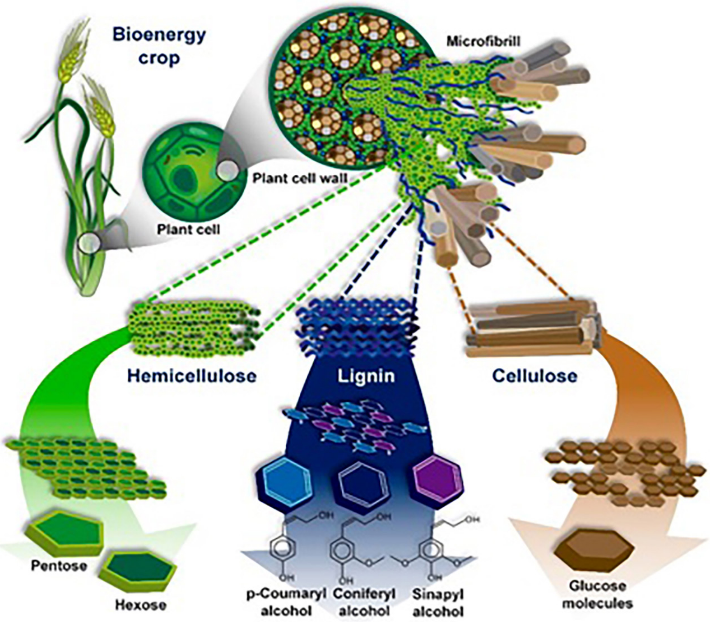


Fig. 2. Structure of lignocellulosic biomass [15]



The proportions of basic components differ depending on a type of the lignocellulose material (Tab. 1). The composition of biomass is significant as it affects the productivity of biofuels and their energetic efficiency [22].

Lignocellulosic biomass is one of the most abundant sources of bioenergy. Its global annual energetic potential is estimated at 100–270 EJ [16]; however, it is a material resistant to enzymatic hydrolysis what is determined by the structural factors, including crystallinity of cellulose and degree of its polymerisation, size of pores, volume and also, chemical factors, including the composition and the content of lignin, hemicellulose and acetyl groups [18].

Bioethanol from lignocellulosic biomass may be obtained by the thermo-chemical or biochemical conversion. In thermo-chemical process, the raw material is subjected to gasification and gas obtained from the synthesis is converted into ethanol with the application of *Clostridium ljungdahlii* bacteria. Production of cellulosic bioethanol by biochemical method is a complex, multistage process and it covers *inter alia*, preliminary treatment of raw materials, enzymatic hydrolysis, sugar fermentation and recovery of ethanol [23].

Table. 1. Composition of different lignocellulosic substrates [15]

Lignocellulosic substrates	Composition (% dry basis)		
	Cellulose	Hemicellulose	Lignin
bamboo stem	43.04	22.13	27.14
birch	40.1 ± 0.6	17.5 ± 0.2	24.2 ± 0.1
corn cob	42.0 ± 0.1	45.9 ± 0.9	2.8 ± 0.2
corn stalk	36.4 ± 0.1	30.3 ± 0.1	6.9 ± 1.4
corn stover	42.21	22.28	19.54
corn straw	49.3 ± 1.8	28.8 ± 1.4	7.5 ± 0.4
cotton stalk	41.6 ± 0.5	23.6 ± 0.4	23.3 ± 0.7
eucalyptus	52.07 ± 2.6	24.51 ± 1.1	25.2 ± 1.1
empty fruit bunch	34.9	26.64	31.1
giant reed	41.5 ± 2.6	20.5 ± 0.6	18.4 ± 1.4
grass	47.12 ± 3.2	36.01 ± 3.17	11.55 ± 0.3
maize straw	38.33 ± 0.8	29.76 ± 1.35	3..82 ± 0.5
meadow grass	41.28 ± 5.3	28.14 ± 3.2	30.14 ± 7.9
Miscanthus	36.3 ± 2.1	22.16 ± 1.9	22.55 ± 2.5
oat straw	35.0	28.2	4.1
oil palm empty fruit bunch	38.5 ± 1.9	26.1 ± 1.1	11.6 ± 1.6
pinewood	38.2 ± 0.3	24.1 ± 0.7	34.4 ± 0.3
poplar	46.0 ± 0.1	16.7 ± 0.1	26..6 ± 0.3
rice hulls	36.0	12.0	26.0
rice straw	37.8 ± 0.2	29.6 ± 0.7	14.8 ± 0.4
rye straw	36.5 ± 0.1	not reported	21.3 ± 0.1
sawdust waste	31.5 ± 1.3	26.1 ± 2.1	24.9 ± 1.7
sorghum straw	26.93 ± 1.2	32.57 ± 1.9	10.16 ± 1.8
spruce	24.7 ± 0.2	10.2 ± 0.1	35.0 ± 0.3
sugarcane bagasse	46.1 ± 0.7	20.1 ± 0.9	20.3 ± 0.6
sunflower stalk	34 ± 0.6	20.8 ± 0.8	29.7 ± 0.6
water hyacinth	36.84 ± 0.8	27.7 ± 0.2	10.7 ± 0.4
wheat straw	43.4	26.9	22.2
willow sawdust	35.6 ± 0.9	21.5 ± 0.9	28.7 ± 0.2

Preliminary treatment of lignocellulosic biomass

The basic problem consists in effective separation of sugars from lignocellulosic biomass. The preliminary treatment changes its structure and owing to this fact, the conversion of polysaccharides to fermentable sugars is possible [18]. The significant factor includes first of all removal of lignin, limiting the cellulose and hemicellulose hydrolysis in a great degree. The discussed processes utilize the methods which affect not only the content of lignin but also change a crystalline nature of cellulose and decrease the size of the particles of the initial material what results in rise of the level of lignocellulosic biomass digestibility [24]. After the preliminary treatment, cellulose is exposed, more available for cellulases and is hydrolysed quicker than that the untreated one [25].

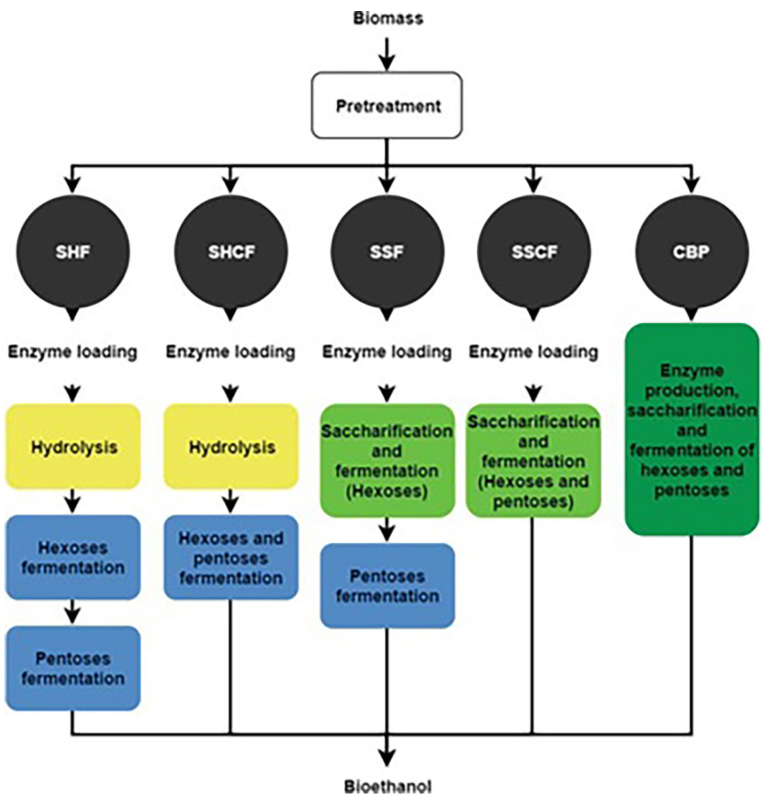
The features of ideal pre-treatment process:

- Obtaining of cellulose substrate sensitive to enzymatic hydrolysis,
- Minimum degradation of sugars or carbohydrates,
- Generation of minimum inhibitors and compounds toxic for microorganisms responsible for ethanol fermentation process,
- Low energy consumption in the process,
- The conditions of the process are favourable for lowering of the capital and operating costs,
- Inexpensive and recyclable chemicals used in the pre-treatment,
- The possibility of treating different types of biomass,
- Generation of by-products from lignin and hemicellulose, being suitable in various other industrial sectors,
- Scalability of the process, enabling its application in commercial purposes,
- A small negative impact on the environment [16].

Basic methods of preliminary treatment include as follows:

- Physical methods – they are employed with the aim to destroy a crystalline structure of cellulose, to decrease the size of the particles and to increase the area of surface of the raw material; they do not affect the chemical composition of the raw material; cellular walls are not subject to degradation; the significant structural changes do not occur,
- Chemical methods – they utilize chemical reagents, generally at the increased temperatures; they may cause a removal of hemicellulose by its dissolving or hydrolysis, removal of lignin (delignification) by destroying of the structures (depolymerisation and dissolving), liquefying of cellulose and preliminary hydrolysis [16],
- Physico-chemical methods – they employ water vapour explosion, combination of water vapour explosion and alkaline methods of the preliminary treatment of raw material in liquid anhydrous ammonia in the conditions of high temperature (90–100°C) and high pressure (1–5.2 MPa), application of CO2 which during water vapour treatment generated carbonic acid, facilitating hemicellulose hydrolysis: they use SO2 and acids at low temperatures to a partial dissolving of cellulose and catalytic technologies, based upon the oxidation processes [26],

Fig. 3. Processes for the second generation bioethanol production [32]



- Biological methods – they utilize lignolytic potential of certain microorganisms; owing to hydrolytic enzymes (oxidoreductases), microorganisms are able to decompose lignin effectively; the processes of decomposition may be conducted when cultivating microorganisms directly on the lignocellulose raw material or using enzymatic extracts [27].

After the preliminary treatment, the substrate is subjected to enzymatic depolymerisation. It is the most expensive stage due to a high cost of enzymes. In the obtained hydrolysate, C-5 and C-6 sugars are found which may be subjected to fermentation with generation of bioethanol [11]. In the process of ethanol fermentation, we may employ various modifications (Fig. 3), including separation of the process of enzymatic hydrolysis and fermentation [28] and run each of them separately in the optimal conditions (SHF method – separate hydrolysis and fermentation). Hydrolysis and fermentation may be conducted also together (SSF – simultaneous sacharification and fermentation). Sugars generated during hydrolysis are simultaneously fermented to ethanol. The studies confirm the effectiveness of the discussed technology of the 2nd generation bioethanol production from lignocellulose wastes [29]. In SSCF method (simultaneous sacharification and co-fermentation) not only sacharification and fermentation of hexoses but also fermentation of pentoses takes place [30]. In SHCF method (separate hydrolysis and co-fermentation), the processes of hydrolysis and fermentation run separately, whereas the fermentation of hexoses and pentoses occurs simultaneously. In CBP (consolidated bio-processing) method, we may combine production of enzymes, enzymatic sacharification and fermentation in one stage [31].

Lignocellulosic bio-refineries

Processing of lignocellulosic biomass in the so-called bio-refineries is economically justified; apart from the fuel, we may also obtain valuable bio-chemicals and biomaterials for application in other sectors of industry (Tab. 2). To produce them, the parts of biomass being not used directly in ethanol production, are first of all employed.

The high-value by-products may be obtained from the lignin, being separated in the earlier stage of the process (Fig. 4) and from hemicelluloses and C5 sugar subjected to a separate fermentation (Fig. 5). Additionally, some different energy carriers may be also produced (Fig. 6).

In 2017, there were 224 biorefineries acting in Europe, however only 43 were the second generation bio-refineries, utilizing, *inter alia*, non-consumption plants and bio-wastes. In the

Table. 2. Lignocellulosic biorefinery products [11]

Biofuels	Bioenergy	Food products	Biochemicals	Biomaterials
biodiesel, bioethanol and biomethane	steam power, electricity steam, syngas, heat, charcoal and lignin	sugar and substitutes, proteins, amino acids, gluten, protective colloids thickeners, emulsifiers and stabilisers	simpler hexose and pentoses, and their degradation products such as 5-hydroxy methyl furfural, glycerol, agrochemicals, fertilisers, sorbitol, phenols, coloured compounds, solvents, omega-3 fatty acids and biosurfactants	pulp and papers, PHB, activated carbon, bioplastics, bio-based epoxy, resin, cement, bioadhesives, bio-based polymers, bio-nanocomposites, etc.

Fig. 4. Conception of "lignin-driven" bioethanol based biorefinery [33]

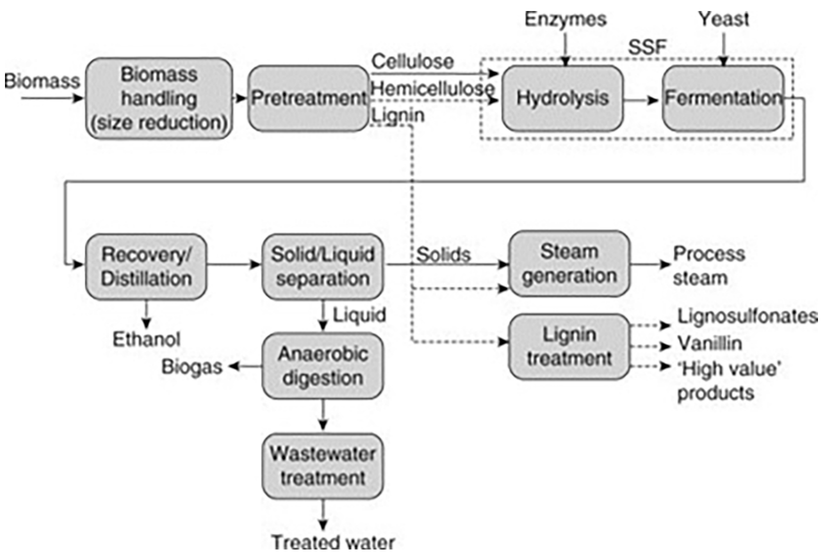


Fig. 5. Conception of "C5-driven" bioethanol-based biorefinery [33]

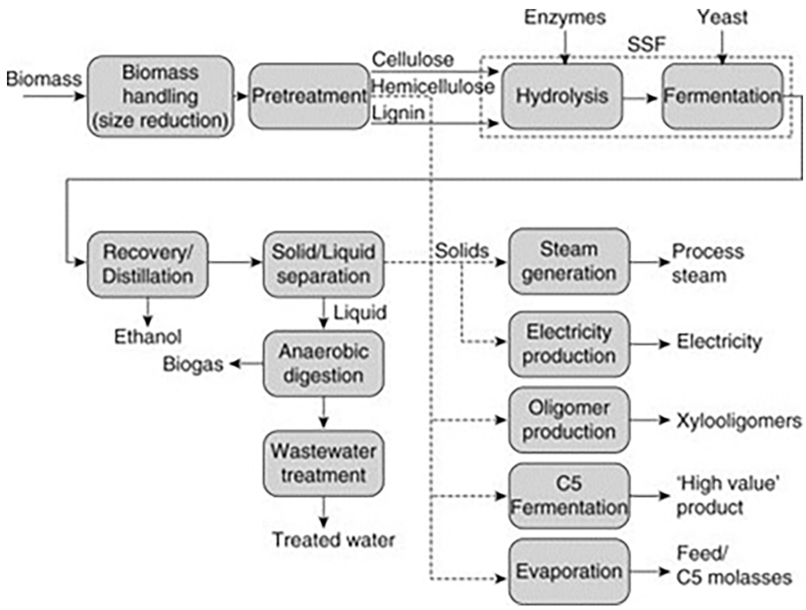
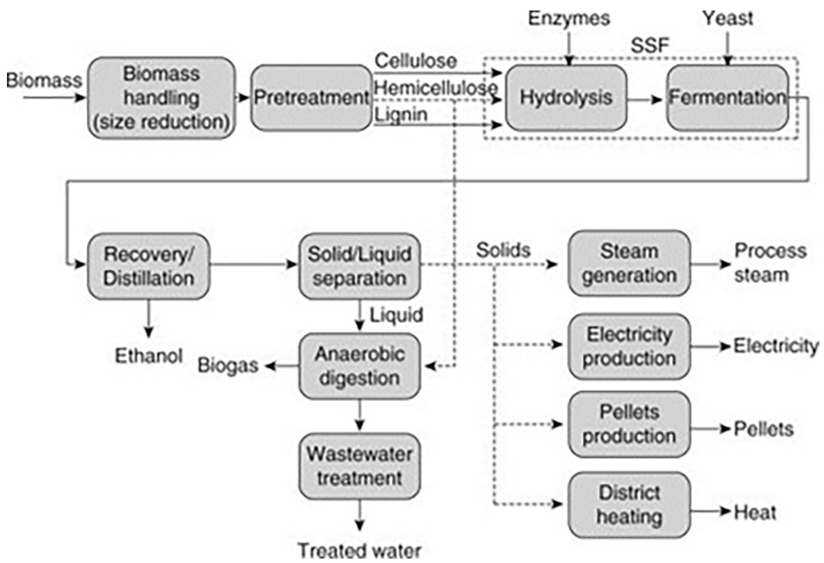


Fig. 6. Conception of "energy-driven" bioethanol-based biorefinery [33]



previous years, the EU finances a lot of the projects connected with the biorefining industry (Tab. 3), concerning the utilization of various fractions of biomass in the efficient and sustainable way [34]. Production of ethanol from cellulose materials is a promising technology which may help in energetic diversification and in decarbonisation of transport sector. Cellulosic bioethanol allows not only GHG emissions but has also a low ILUC coefficient connected with the indirect consequences of the changes in land utilization. Moreover, it is subjected to mechanism of double counting what means that the content of energy in the discussed biofuel is twice calculated in the total aim in respect of renewable energy in transport [35].

It is estimated that the investment costs on the plants of this type are found in the limits between 2570 EUR/kW and 3650 EUR/kW for ethanol production and it is dependent, *inter alia*, on the size of the plant, complexity of technology and location. The costs of raw material are dependent first of all on its type and availability in the site and are estimated at 10–20 EUR/MWh (50–100 EUR/tonne of dry matter) we should also add the costs of enzymes necessary for the hydrolysis process of order 15–30 EUR/MWh of the product and the remaining operating costs in amount of 13–18 EUR/MWh connected with the labour costs and purchase of media. As the discussed technologies are found in the stage of commercialization, there is a possibility of lowering the costs (Tab. 4) via optimization of the processes, integration with other technologies being advantageous for improvement of the effectiveness of the obtained by-products [36].

There are already a few plants in the world which produce cellulose ethanol in the commercial scale, e.g. Borregaard Industries AS in Norway (efficiency 16 kilo tonnes), Raizen Energia (36 k tons) and GranBio (65 k tons) in Brasil and POET-DSM Advanced Bio-fuels in the USA (75 k tons) [35].

Table 3. The selected projects, financed by the EU concerning utilization of lignocellulosic raw materials in biorefineries [34]

Project name	Biorefinery feedstock	Country coordinated in	Period	Total cost (€)
AgriChemWhey	Byproducts from dairy processing	Ireland	2018–2021	29 949.323
GRACE	Miscanthus or hemp varieties from marginal lands	Germany	2017–2022	15 000 851.21
SmartLi	Kraft lignins, lignosulfonates, and bleaching effluents	Finland	2015–2019	2 407 461.25
BIOSKOH	Lignocellulosic feedstock	Italy	2016–2021	30 122 313.75
BARBARA	Agri and food waste	Spain	2017–2020	2 711 375
AgriMax	Agri and food waste	Spain	2016–2020	15 543 494.56
PULP2VALUE	Sugarbeet pulp	Netherlands	2015–2019	11 428 347.50
GreenSolRes	Lignocellulosic residues or wastes	Netherlands	2016–2020	10 609 637.01
Dendromass4Europe	Dendromass on marginal land	Germany	2017–2022	20 442 318.75
YLFEED	Wood residues	France	2017–2020	14 976 590
GreenProtein	Vegetable residues from packed salad processing	Netherlands	2016–2021	5 546 519.99
PROMINENT	Cereal processing side streams	Finland	2015–2018	3 103 897.50
FIRST2RUN	Cardoon from marginal lands	Italy	2015–2019	25 022 688.75
Zelcor	Lignocellulosic residues from ethanol production, lignins dissolved during pulping process, and lignin-like humins formed by sugar conversion	France	2016–2020	6 710 012.50
STAR4BBI	Lignocellulosic feedstocks from forests and agriculture	Netherlands	2016–2019	995 877.50
BIOrescue	Wheat straw and agro-industrial waste	Spain	2016–2019	3 767 587.50
OPTISOCHEM	Residual wheat straw	France	2017–2021	16 376 816.83
US4GREENCHEM	Lignocellulosic feedstock	Germany	2015–2019	3 803 925
FUNGUSCHAIN	Mushroom (*Agaricus bisporus*) farming residues	Netherlands	2016–2020	8 143 661.25
POLYBIOSKIN	Food waste	Spain	2017–2020	4 058 359.38
ValChem	Woody feedstock	Finland	2015–2019	18 502 703.25
LIBBIO	Andes lupine from marginal lands	Iceland	2016–2020	4 923 750

Table 4. Potential costs of cellulosic ethanol production after reductions [36]

Process	**Costs, EUR/MWh**					
	Low cost feedstock: 13 EUR/MWh			High cost feedstock: 20 EUR/MWh		
	SGAB*	Future costs		SGAB*	Future costs	
		Pessimistic	Optimistic		Pessimistic	Optimistic
Investment cost EUR/kW output	(SGAB Low) 2,570	(SGAB-25%) 1,928	(SGAB-50%) 1,285	(SGAB High) 3,650	(SGAB-25%) 2,738	(SGAB-50%) 1,825
	Total production costs (EUR/MWh)					
Capital	42	32	21	60	45	30
Feedstock	33	33	33	50	50	50
Operating costs	28	25	22	48	27	24
Total	103	90	76	158	122	104

** Sub-Group on Advanced Biofuels*

Summing up

Lignocellulosic biomass is a material which may be successfully utilized as a raw material for production of the second generation bioethanol. Its manufacture is not dangerous to the food production and has a positive impact on the environment. The application of cellulose ethanol reduces energy consumption from traditional carriers so it is favourable for limitation of greenhouse gases' emission. Its production is, however, purposeful in biorefining processes where – apart from the biofuels itself – we may obtain many additional products, electric energy and heat. They constitute the value added and have a favourable effect on economic balance of the whole process.

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