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Analysis of Properties of Synthetic Hydrocarbons Produced Using the ETG Method and Selected Conventional Biofuels Made in Poland in the Context of Environmental Effects Achieved

Małgorzata Krzywonos[*], *Karol Tucki*[**], *Janusz Wojdalski*[**], *Adam Kupczyk*[**], *Michał Sikora*[**]
[*]*University of Economy, Wrocław,*
[**]*Warsaw University of Life Sciences*

1. Introduction

For many years, the Polish energy mix has been based on traditional, non-renewable carriers of primary energy, in particular, brown coal and hard coal. End use of fuels in transport is also dominated by non-renewable fuels, that is, Diesel oil and unleaded petrol. The climate and energy policy of the European Union, the so-called 3x20% + 10% package, indicates activity aimed at promotion of renewable energy sources both in power engineering and in transport. According to provisions of Directive 2009/28/EC, until year 2020, the share of biofuels in total fuel consumption is to reach at least 10% in terms of energy (Krzywonos et al. 2015). The new long-term objectives for the climate and energy policy have been agreed and accepted in October of 2014 by the European Council. They require a 40% reduction in CO_2 emission and at least a 27% share of energy from renewable sources (RES) at the EU level in final energy consumed by the end of year 2030. The new energy policy took into account the motion for amendment of directive on renewable energy sources and sustainable use of bioenergy. The Community activities include incentives to use financial instruments to support development of new RES generation capacity and promotion of cooperation between member states.

In April 2015, the European Parliament and the Council agreed to amend, until the end of year 2020, the directive on fuel quality and the RES directive in order to take into account the effects of indirect change in the mode of land use, caused by increase in specific crops, used for biofuel production. The following assumptions have been made in the new legal framework:

- The level of maximum 7% share of biofuels made of edible raw materials, which are to be designated for achievement of the objective of 10% share of energy from renewable sources in transport until 2020;
- A target estimated level of 0.5% is to be introduced with regard to advanced biofuels; and
- The Commission will be obliged to take into account the effects of indirect change of land use (ILUC) through inclusion of emission indicators in the reports (European Commission 2015).

The components, listed above, have been taken into account in the prepared draft of amendment of the act on amendment of the act on biocomponents and liquid biofuels of August 31st, 2016, which is now in the course of social consultations (Ministry of Energy, 2016).

Increase in the share of renewable energy in the transport sector is implemented mainly through introduction of transport biofuels on the market, primarily by mixing biocomponents with traditional fuels. For more than 10 years, a well developed technical infrastructure has been present in Poland, allowing for ethanol production, which is translated to very high production capacity for bioethanol, amounting nearly to 750 million dm^3,while the real consumption level remained relatively low, at the level of 24.4%. Overall generation capacity for biocomponents, bioethanol and methyl esters in Poland exceed 2 bln l/year (Borowski 2014, Krzywonos et al. 2015, Antczak et al. 2016).

In the recent years, a number of amendments have been made to the legislation (Act of 2015) regulating the domestic sector of transport biofuels, aimed at reducing energy consumption and emission levels of production processes, as well as partial replacement of the raw materials used.

These amendments concerned, among other things, introduction of the Sustainable Development Criteria (*SDC*) with regard to minimum levels of CO_2 emission reduction, which must be met by all biofuels and biocomponents marketed.

At present, many production plants have had to undergo detailed energy audits of their production lines in order to optimize primary energy, heat and electricity consumption. Modernization and/or modification tasks were based largely on application of natural gas as the process fuel, which is processed by highly efficient cogeneration. These activities are based mainly on the previously identified (e.g. Borowski et al. 2014) problems with regard to sustainability, including reduction of CO_2 emission for selected biofuels made of raw materials originating from the food industry – in particular, these apply to bioethanol produced in two phases (distillery – dehydration facility).

At present, the minimum levels are 50% starting from 2018 and 60% for biofuels generated in new installations (Act of 2015). All of these additional regulations are aimed at stimulating and accelerating implementation of the technology for production of 2nd generation biofuel (advanced biofuels), which are made of non-food, waste or post-production biomass (Act of 2015). Such biofuels include, for instance, bioethanol, made in the process of fermentation of lignocellulosic biomass (Wang 2012,Wilk 2015, Munoz 2016), originating from waste from baking industry, as well as biodiesel, originating, among others, from fat processing waste.

These biofuels are characterized by much higher (more beneficial) levels of CO_2 emission reduction (Wang 2012), and they do not compete with food crops (Munoz 2016, Krzywonos 2014). It should be noted that legislation on marketing of 2nd generation biofuels have been subject to substantial fluctuations lately, in particular, with regard to their eligibility for double counting for the purpose of achievement of the National Indicative Target (*NIT*). A substantial part of the production capacity of bioethanol can be still potentially used, in particular, in the case of more favorable market conditions of increased demand. This leads to the conclusion that the Polish market of bioethanol seems to have a great potential for unconventional technologies of ethanol processing to synthetic hydrocarbons, in particular, ETG (Ethanol to Gasoline). Moreover, it can be predicted that development of this technology will lead to a positive change on the market of bioethanol and result in increased demand for ethyl alcohol produced by Polish distilleries (Krzywonos 2015). One of such new ETG installations will soon start to produce advanced biofuels in the process of catalytic hydrogenation of ethanol, obtaining synthetic hydrocarbons (Book EkoBenz 2016). The installation will allow, on the industrial scale (22 500 Mg/year), of processing of ethanol to receive synthetic hydrocarbons of parameters consistent with the present quality standards for motor fuels to be marketed.

Alcohols were used as motor fuels already in the 19th century. Among alcohols, ethanol is most popular, and lately, it has been presented as a renewable, biomass-based, environment-friendly motor fuel for spark-ignition engines (Faber 2011, Rodriguez-Anton et al. 2016, Sikora 2016).

Ethanol is a promising alternative for biomass fuel thanks to its chemical properties as well as the fact that it is biodegradable (He et al. 2003). Addition of ethanol and/or ETBE to gasoline may change some fuel properties and gas emission, mechanical capacity and reliability of the engine, at the same time warranting maintenance of good performance standards (Rodriguez-Anton et al. 2016) and increasing the octane rating of mixed fuels and reducing distillation temperature (He et al. 2003, Sikora 2016).

Catalytic ethanol transformation to synthetic hydrocarbons in the ETG technology (Fig. 1) can be divided into three stages: ethanol dehydration to ethylene, secondary reactions (oligomerization of ethylene), production of aromatic compounds/ paraffin through hydrogen transfer (HT) (Sun 2014).

Research on conversion of bioethanol to dimethyl esthers has been conducted, among others, by Nazimek et al. (2015a,b) using copper ion-exchanged zeolite catalysts. The properties of hydrocarbons obtained in the ETG process have been presented in Tabela 1.

Dimethyl ether (DME) has been considered one of the substitutes of Diesel fuel. This has been due to development of cheaper and simpler technologies of its production. The reason for it use as diesel fuel include low self-ignition temperature and high cetane number, which improve the engine performance qualities. Moreover, in comparison with diesel fuel, emission levels of greenhouse gases and other harmful substances have been reduced. Thanks to use of bioethanol for DME production, it can be used as biofuel (BioDME) (Nazimek 2015).

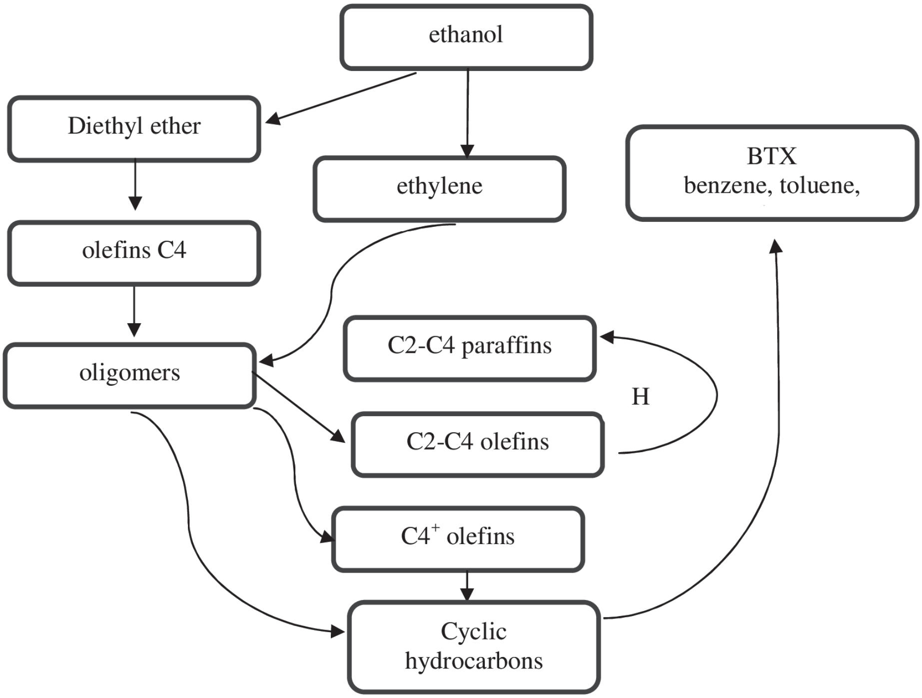

Fig. 1. Conversion of ethanol to gasoline (ETG)
Rys. 1. Proces konwersji etanolu do benzyny (ang. ethanol to gasoline ETG)
Source: Own compilation on the basis of (Viswanadham 2012)

Refineries will be forced to produce increasingly environment-friendly fuels. Adding of synthetic hydrocarbons seems to be an ideal/ advantageous solution. If attractive unit price can be attained for synthetic hydrocarbons, ethanol can be completely replaced as a biocomponent. In production of ethanol of 1st generation, the share of the cost of purchase of enzymes in relation to total production cost of alcohol is not significant, while in production of 2nd generation ethanol, the enzyme cost can reach even 15% of total cost. Thus, the present efforts in the field of 2nd generation ethanol are aimed at increasing enzyme performance and activity. At present, enzyme cost for production of 2nd generation ethanol, according to ABENGOA company, are estimated to range between USD 0.10 and 0.13 per 1 dm^3 of ethanol (Ramos 2016).

Table 1. Chemical composition of synthetic carbohydrates obtained using ETG method
Tabela 1. Skład chemiczny węglowodorów syntetycznych uzyskanych metodą ETG

Chemical composition	Synthetic hydrocarbons after fractioning at capacity of 80%	Synthetic hydrocarbons obtained using ETG method (raw material: ethanol)	Synthetic hydrocarbons obtained using ETG method (raw material: methanol)	Synthetic hydrocarbons obtained using ETG method (raw material: butanol)	Nazimek and Niećko (2010)	Quality requirements for motor gasolines (Resolution of 2008)
MeOH, % vol/vol	<0,17					3
MeOH, % vol/vol	<0,17					5
ETBE Ethyl tert-butyl ether, % vol/vol					22.7	
Oxygen % [m/m]	<0,17	0.42	0.59	0.15	3.28	2.7
Benzene, % vol/vol	<0,1	0.08	0.00	0.01	–	1.0
Olefin type hydrocarbon content, % vol/vol	4.4	7.5	15.8	10.1	8.57	0-18,00
Saturated compounds, % vol/vol		59.8	58.3	31.4	52.35	
Aromatic hydrocarbon content, % vol/vol	28.5	30.7	23.6	26.8	16.35	0-35
Xylene, % vol/vol		4.9	2.6	1.9	-	
RON (Research Octane Number)		94.8	86.5	84.9	107.9	95-

Source: Own compilation

An economically efficient solution could be withdrawal of excise tax on ethanol for energy production purposes. This could substantially increase the attractiveness of ethyl alcohol as a raw material for synthetic gasoline production. At present, excise tax for motor gasolines and products of mixing of these with biocomponents is PLN 1540 / 1000 l. The rate for diesel fuels with CN code is PLN 1171/ 1000 l. In relation to biocomponents constituting independent fuels (CN code is of no significance), which meet the specific quality requirements, the rate of PLN 1171/1000 l is also applicable. For other motor fuels, it is PLN 1797/1000 l. The excise tax for ethyl alcohol is even higher, amounting to PLN 5704 /100 l of 100% ethanol (Resolution of 2015). Such high financial burden leads to a substantial slowdown in research on potential uses of alcohol as an energy source.

At present, a very significant factor, which determines the possibility of marketing of innovative, advanced biofuels and biocomponents for the *NIT* is compliance with the requirements of the *SDC.* These criteria reflect the minimum acceptable levels of the environmental (ecological) effect obtained as a result of use of biofuels and they are expressed as reduction in CO_2 emission. In this work, we focused on comparative analysis of selected conventional 1st generation biofuels and the advanced biofuel (synthetic biohydrocarbons) obtained using ETG methods with regard to environmental effect.

The aim of this work was to conduct a comparative analysis for synthetic biohydrocarbons produced using the ETG method and selected conventional biofuels (of 1st generation), made of food components, from the perspective of the environmental effect achieved, expressed as CO_2 emission reduction.

2. Analysis methodology

The comparative analysis was conducted for selected biofuels produced in various regions of Poland. The research was conducted using the LCA (Life Cycle Assessment) technique and the BIOGRACE 4.0 d procedure/ tool. It has been approved by the European Commission (BioGrace GHG 2016) as compliant with the greenhouse gas emission assessment methodology in association with production processes and use of transport fuels, biofuels and bioliquids, specified in Directive 2009/28/EC. The tool is generally available and used for determination of

CO_2 emission reduction levels and compliance of transport biofuel production systems with the sustainable development criteria (Biograce GHG, 2016). Moreover, within the framework of the analysis conducted, it is possible to assess energy crop biomasses designated for biofuel production. As for processing of biomass for energy production purposes, the tool allows for calculation of agricultural emissions, as well as emissions and emission reductions in the full life cycle of plants generally used for bioethanol and biodiesel production, as well as for determination of direct impact of the value of a given parameter of the unit biomass and/or biofuel production process on the final emission value, expressed in relation to the functional unit applied (1 MJ of biofuel) throughout the full life cycle.

Research was conducted using detailed data, typical for the selected biofuel production technologies, including in particular: i. bioethanol made of corn (1 phase method), ii. synthetic biohydrocarbons using the ETG method (advanced biofuel). As a supplement for missing, unavailable data, standardized indicator values were used, typical for selected links of the production chain. In order to reflect the cases analyzed more precisely, modified indicator values, typical for Poland, were also applied, e.g. with regard to emission from nitrogen-based fertilizers.

2.1. Analysis of parameters of synthetic biohydrocarbon production using ETG method (case 1)

The production process was analyzed using the LCA (Life Cycle Assessment). The process of production of synthetic biohydrocarbons consists of the following unit processes:

1) Production of ethanol, including
   - Plant cultivation,
   - Transport of grains/ plants to the plant
2) Transport of ethanol to the plant for production of synthetic biohydrocarbons using the ETG method,
3) Production of synthetic biohydrocarbons,
4) Transport to production site.

A simplified diagram of production for the analyzed life cycle of the process of production of synthetic biohydrocarbons using the ETG methods, divided into unit processes, has been presented in Figure 2.

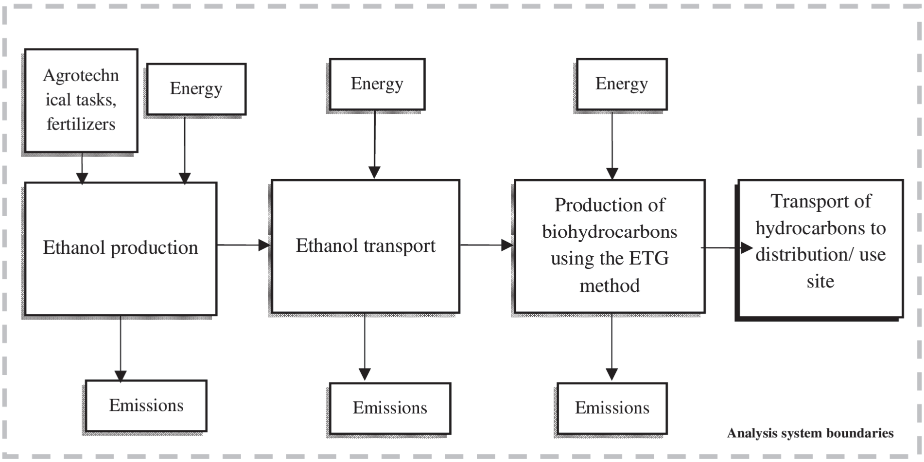

Fig. 2. Scheme of the life cycle of the process of synthetic biohydrocarbons produced by ETG
Rys. 2. Schemat cyklu życia procesu wytwarzania biowęglowodorów syntetycznych metodą ETG
Source: own compilation

Indicated in figure 2 are the main material and energy flows in the life cycle. For the sake of simplification, a detailed division of the ethanol production process has not been presented – it has been explained in the further part of the text (see chapter 2.3), however, the authors indicate that in the final results of analysis of the ecological effect, its energy consumption and the resulting emission values have been taken into account. The process of production of synthetic biohydrocarbons by ETG is much more complex than the processes of production of conventional biofuels. This is mainly due to the possibility of replacing a part of the main stream of substrate (pure ethanol) with substances constituting waste and/or byproducts of alcohol production processes. This has a very positive impact on the final energy and emission balance of the production process. An overall block diagram of the manufacturing process for synthetic biohydrocarbons has been presented in Fig. 3.

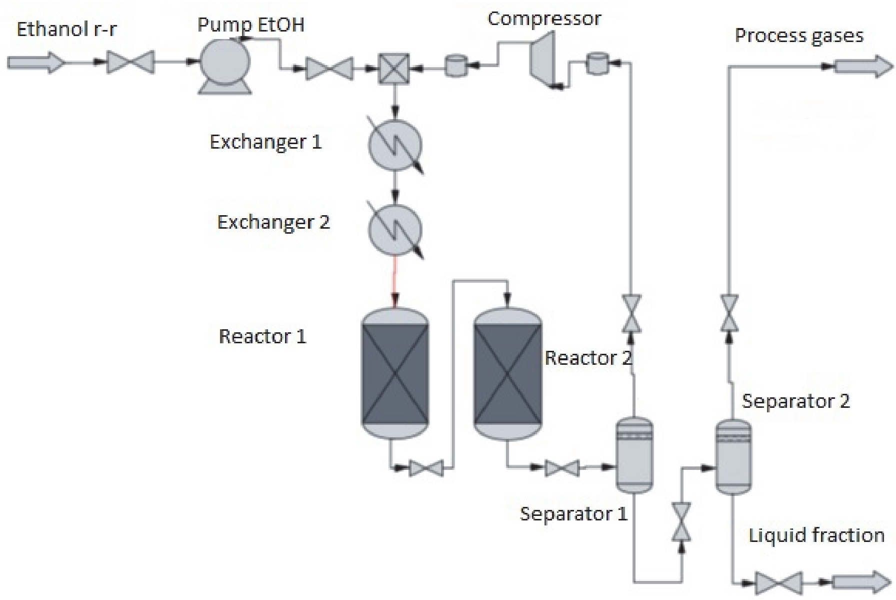

Fig. 3. Block diagram of the process of manufacturing of synthetic biohydrocarbons by ETG method
Rys. 3. Blokowy schemat poglądowy procesu wytwarzania biowęglowodorów syntetycznych metodą ETG
Source: own compilation on the basis of materials of Ekobenz Sp. z o.o.

In this analysis was taken the production of biohydrocarbons (the drained ethanol), which is the low wine produced in the 1-phase method from beet molasses, in the plant where the source of technological heat is natural gas, which is burned in the high-efficiency steam generators (without cogeneration).

2.2. Analysis of parameters of the process of bioethanol production from corn using the 1-phase method (case 3)

The life cycle of bioethanol production from corn using the 1-phase method consisted of three main unit processes:
1) Corn cultivation
2) Transport of corn grains to bioethanol production plant,
3) Production of bioethanol at the plant using the 1-phase method,
4) Transport of bioethanol to the final use/distribution site.

The block diagram of life cycle of bioethanol production from corn has been presented in figure 4.

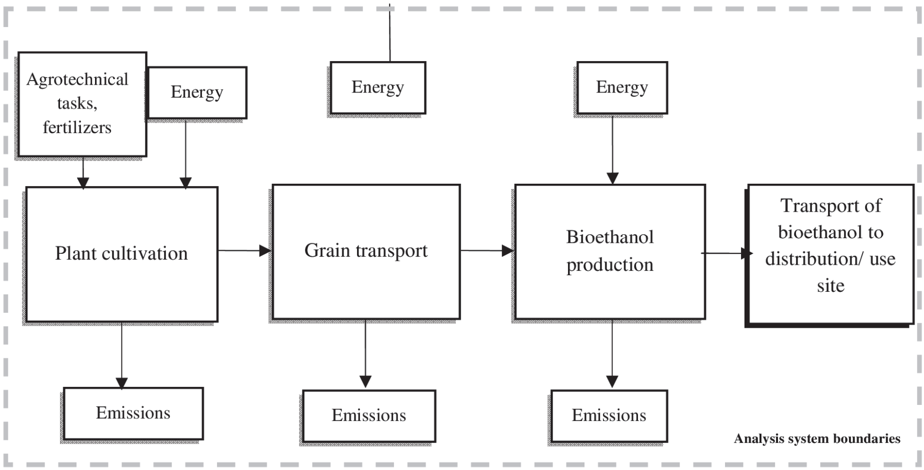

Fig. 4. A block diagram of the life cycle manufacturing process of bioethanol of corn in a 1-phase process
Rys. 4. Schemat blokowy cyklu życia procesu wytwarzania bioetanolu z kukurydzy metodą I-fazową
Source: own compilation

It presents an outline of a block diagram of the life cycle of the process of bioethanol production from corn using a 1-phase method, including the main unit processes and the key input and output streams of mass and/or energy. Bioethanol was produced using the pressure method with enzymatic hydrolysis of substrates. The process fuel was natural gas, combusted in highly efficient steam generators (processing efficiency at the level of 85%). It was assumed that the total transport distance for individual raw materials and products would be 350 km. The production scale at the plant under concern was 30 000 dm^3/24h. A block diagram of the production process has been presented in Fig. 5.

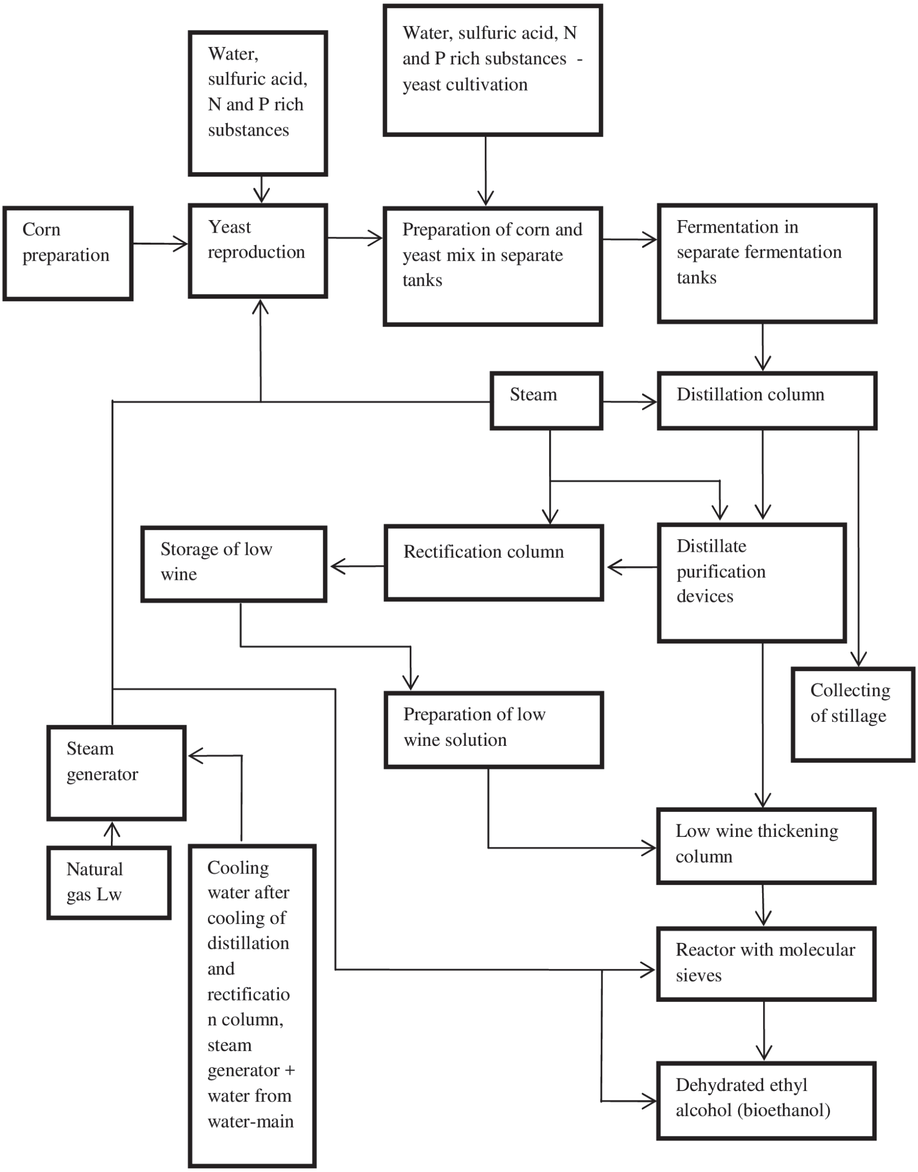

Fig. 5. Block model (technological path)of bioethanol producing-plant from corn by single-phase method
Rys. 5. Model (ścieżka technologiczna) zakładu produkującego bioetanol metodą jednofazową z kukurydzy
Source: Kupczyk, et al. (2016).

3. Analysis results

Table 2 presents results of the comparative analyses conducted, environmental effects (CO_2 emission reduction for synthetic biohydrocarbons produced using the ETG method and conventional biofuels of the 1st generation).

Table 2. Compilation of the results of the calculations for bioethanol production from corn by using single-phase method and and advanced biofuels (ETG)
Tabela 2. Zestawianie wyników obliczeń dla przeanalizowanych procesów wytwarzania biowęglowodorów syntetycznych metodą ETG oraz konwencjonalnych biopaliw 1. generacji

SYNTHETIC BIOHYDROCARBONS (case 1)	
Emission value for transport of ethanol to production plant [g CO_2eq/MJ]	< 1
Emission value for the production process [gCO_2eq/MJ]	< 28
Emission reduction for the production process calculated using BIOGRACE [%]	**< 60**
1-PHASE BIOETHANOL (case 3)	
Quantity of bioethanol produced, dm^3	30,000.00
Quantity of bioethanol produced, MJ	649,874.40
CO_2 emission in corn cultivation, g/MJ of ethanol	26.08
CO_2 emission in cultivation of plants to be used for bioethanol production, g/kg of corn	179.85
CO_2 emission in bioethanol production, g/MJ of ethanol	60.30
CO_2 emission in transport throughout the entire life cycle in total, g/MJ of ethanol	1.9
Total emission after allocation, g/MJ of bioethanol	49.0
CO_2 emission for bioethanol production [%]	**41,5-43**

Source: own compilation and Kupczyk et al.(2016)

The results obtained for the synthetic biohydrocarbon production process (case 1) indicate the environmental effect values above 60% of CO_2 emission reduction throughout the entire life cycle. These are also the highest among all transport biofuels analyzed. This indicates that the final energy consumption for this production process is the lowest, in comparison with the remaining cases. The main factors that determine the above correlations include *i.* use of waste and/or zero emission semi-finished products as a significant part of the substrate feed, *ii.* low losses in energy transmission at the plant, *iii.* high efficiency of processing of primary energy carriers to heat.

For the case of bioethanol made of corn, they indicate the environmental effect and emission level at ~49 g CO_{2eq}/MJ of bioethanol, which is sufficient to achieve CO_2 emission reduction at the level of 41,5-43%. This means that the plant meets the defined criteria for CO_2 emission reduction, and bioethanol produced at the plant can be marketed. At the same time, it should be noted that at present, the biofuel does not meet the reduction threshold of 50%, which is to be applicable starting from January 1st, 2018. At the same time, it has been indicated that such installation, after the loss limitation and energy consumption optimization processes, may meet the required criterion of CO_2 at the level of 50%. The results presented refer to the works of Kijeński (2007), Gąsiorek and Wilk (2011), Manzetti & Andersen (2015, 2016).

4. Summary

Hydrocarbons produced using the ETG method can be used as a biocomponent in fuels for spark-ignition engines. The synthetic hydrocarbons obtained, from chemical point of view, meet the quality requirements for motor fuels used in vehicles with spark-ignition engines. The environmental effect values achieved (above 60%) indicate that this modern fuel may successfully be used in the future as an innovative renewable fuel. In comparison with conventional biofuels, generally used on the Polish market of biofuels of the 1st generation, the environmental effect achieved is higher by more than 20 percentage points. High values of CO_2 emission reduction are, among other things, attributable to use of substrates consisting of zero emission waste and/or semi-finished products and high efficiency of processing of primary energy. Further works on development and implementation of ETG technology may bring further improvement in optimum energy consumption, enhancing the environmental effect of CO_2 emission reduction.

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Analiza właściwości syntetycznych węglowodorów wytwarzanych metodą ETG i wybranych konwencjonalnych biopaliw wytwarzanych w Polsce w kontekście osiąganych efektów środowiskowych

Abstract

The aim of the work was to analyze the properties and to compare the processes of production of synthetic biohydrocarbons using the ETG method and the selected conventional transport biofuels produced in Poland, from the perspective of the environmental effect achieved, expressed as CO_2 emission reduction. Research was conducted using the BIOGRACE 4.0 d. method. Within the framework of the research conducted, detailed data ,typical for selected biofuel production methods, was used. The comparative analysis encompassed: *i*. synthetic biohydrocarbons produced using the ETG method (advanced biofuel), *ii*. bioethanol made of wheat (2-phase method), *iii*. bioethanol made of corn (1-phase method).

Streszczenie

Celem pracy było przenalizowanie właściwości oraz porównanie procesów wytwarzania biowęglowodorów syntetycznych wytwarzanych metodą ETG oraz wybranych konwencjonalnych biopaliw transportowych wytwarzanych w Polsce, pod kątem osiąganego efektu ekologicznego, wyrażonego jako redukcja emisji CO_2. Badania przeprowadzone zostały z zastosowaniem metody BIOGRACE 4.0 d. W ramach przeprowadzonych badań zostały wykorzystane dane szczegółowe, charakterystyczne dla wybranych, technologii wytwarzania biopaliw. Analizą porównawczą objęto: *i*. biowęglowodory syntetyczne wytwarzane metodą ETG (biopaliwo zaawansowane), *ii*. bioetanol wytwarzany z pszenicy (metoda II-fazowa), *iii*. bioetanolu wytwarzany z kukurydzy (metoda I-fazowa),

Słowa kluczowe:
bioetanol, redukcja emisji, węglowodory syntetyczne, nakłady energetyczne

Key words:
bioethanol, emission reduction, synthetic hydrocarbons, energy expenditures