

Coupling Ethanol with Synthetic Fuel

Dobiesław Nazimek*, Jerzy Niećko

Department of Environmental Chemistry, Faculty of Chemistry, University of Maria Curie-Skłodowska,
Plac Marii Curie-Skłodowskiej 3, 20-031 Lublin, Poland

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Abstract

This paper presents results of research on the new coupling reaction of ethanol toward higher hydrocarbons (ethanol to gasoline, ETG), which is based on the MTG (methanol to gasoline) reaction focused on selectivity toward ethers, iso- and n-alkanes, and on the activation energy of the ETG reaction. Our paper also reports on the influence of total pressure on the selectivity of the ETG reaction.

Keywords: biofuel, MTG process, ETG process, zeolite catalyst, synthetic fuel

Introduction

There are several traditional methods leading to the production of engine fuels and fuel additives. Fig. 1 presents these methods schematically.

In this paper we will focus on methods leading to high-octane gasoline without giving a detailed description concerning the procedure of adding this biogasoline to gasoline obtained from petroleum. A new way to transform bioethanol to gasoline, called ETG (ethanol to gasoline), is a technological “mutation” of the known MOBIL process based on the coupling reaction of methanol to higher hydrocarbons.

Ethanol as a Component of Gasoline

Essential raw materials for the production of ethanol are: cereals, potatoes, sugar beet-roots, and molasses. The

technology to produce bioethanol is currently advanced, allowing for the possibility of obtaining bioethanol practically from any biomass containing carbohydrates (monosaccharides) and from polycarbohydrates (polysaccharides), namely starch.

The most important steps in the production of bioethanol from polysaccharides are their hydrolysis to monosaccharides and then fermentation to ethanol. The traditional meaning of biogasoline is referred to a mixture of gasoline to which ethanol has been added¹. Oxygen incorporated in ethanol added to gasoline brings about the increase in the octane number of fuels and also decreases the concentration of carbon oxide in the combustion gases by 20-30 vol. %. It also decreases the concentration of unburned hydrocarbons by 10% when compared to the concentration of unburned hydrocarbons emitted by an engine fed with gasoline that does not contain ethanol.

*e-mail: Dobiesław.Nazimek@umcs.lublin.pl

¹Ethanol-containing fuels are designated with the letter “E” (for ethanol) or “M” (for methanol) followed by a number that indicates the volume fraction of alcohol in alcohol-containing fuels. Ethanol (prior to its mixing with gasoline) present in ethanol-containing fuels is denatured and can contain up to 5% hydrocarbons (gasoline-like additives; this is because of the low volatility of ethanol vapor). If a fuel containing 100% ethanol (E100) were used, it would be difficult to ignite an engine at low temperatures. Then the additional and appropriate volume of gasoline is added to denature alcohol in order to obtain the required fuels mixture. Alcohol-containing fuel, E10, is also called “gasohol,” and is a mixture of 10% denatured ethanol + 90% gasoline. E85 is usually called ethanol fuel and has the composition: 70-85% denatured alcohol and 30-15% hydrocarbon fuel.

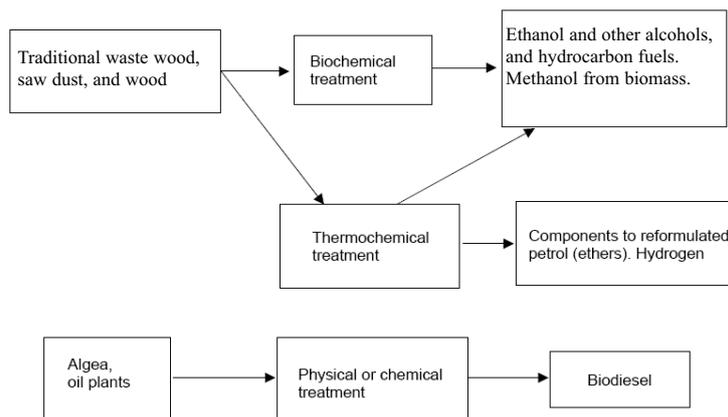


Fig. 1. Traditional methods of biofuel production [1].

However, it is necessary to note that currently and commonly used car engines can run only if the content of ethanol in biofuels is limited to 5%², as in the USA and EU, according to the norm E228. In Poland the content of ethanol in gasoline cannot exceed 5% (see PN-92/C-96025).

Ethanol as a Raw Material for the Production of Gasoline

The case of ethanol production is an essential factor. It mostly depends on both the employed technology and expenditures to pay for raw materials and energy. Depends on the region the price of ethanol varies from 0.9 to 2.60 PLN per kg. For Polish conditions nearly all kinds of raw materials are accessible. The most important raw materials for biofuel production processes can be a variety of non food crops, including:

- oil plants,
- other non food crops (cereals, potatoes, sugar beet root, grass, etc.).

Waste biomass like:

- rural biological wastes (stalks of wheat, corn and wood),
- municipal biological wastes,
- agricultural and food industry wastes (e.g. molasses).

Table 1 presents the potential area for the cultivation of non food crops in select EU countries, including Poland.

The total area of agricultural land in Poland is about 18.5 mln ha, which constitutes about 59% of the area of Poland. The total area of cultivated fields is 12.7 mln ha (68.6% of the total agricultural land) [2].

The greatest shares of cultivated plants in Poland are cereals (including corn) – 71.3%, then potatoes – 10.3% and sugar beet roots – 3.4%. In 1997 the crops of cereals were 25.4 mln tons, potatoes – 20.8 mln tons and sugar beet roots – 15.9 mln tons. Currently, Polish agricultural production is sufficient to cover the buying power of markets,

Table 1. Potential area for cultivation of non food crops for 2005 ([http://www. Biodiesel. de/img99/fig13.htm/](http://www.Biodiesel.de/img99/fig13.htm/)).

Country	Total area of cultivation of non food crops	For biodiesel	For bioethanol
Austria	0.50	0.20	0.30
Belgium	0.20	0.10	0.10
Germany	3.00	1.50	1.50
Denmark	0.50	0.30	0.20
Spain	2.00	1.00	1.00
France	3.00	1.50	1.50
Finland	0.10	0.05	0.05
G. Britain	2.75	1.50	1.25
Greece	0.15	0.05	0.10
Italy	1.00	0.50	0.50
Luxembourg	0.00	0.00	0.00
Ireland	0.50	0.25	0.25
Holland	0.50	0.30	0.20
Portugal	0.50	0.20	0.30
Sweden	0.30	0.05	0.25
Czech Republic	0.60	0.30	0.30
Estonia	0.40	0.20	0.20
Hungary	0.80	0.40	0.40
Poland	3.00	1.50	1.50
Slovenia	0.20	0.10	0.10
Biodiesel	10 mln ha is equivalent to 30-40* mln tons of oil seed crops		
Bioethanol	Biomass		

*Potential weight of crops.

²Because of this, many car manufactures are now producing flexible-fuel vehicles (FFV's) that can safely run on any combination of bioethanol and gasoline, and up to 100% bioethanol. They dynamically sense exhaust oxygen content and adjust the engine's computer system, spark, and fuel injection accordingly (<http://en.wikipedia.org/wiki/biofuel>).

Table 2. Comparison of the amount of different raw materials needed to obtain 1 dm³ of spirit [2].

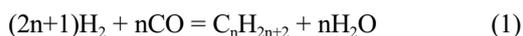
Raw material	Amount required to get 1 dm ³ of spirit [kg]
Potatoes	12.5
Cereals	3.0-3.3
Sugar beet roots	12.5
Molasses	3.3

but some difficulties occur when selling agricultural crops, mainly cereals. Because of this, it is necessary to increase the production of non-food crops that will be potential raw materials for use in the fuel market. Table 2 presents the amounts of raw materials that are required to obtain 1 dm³ of unrefined spirit.

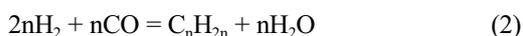
It can be seen from the data presented in Table 2 that molasses and cereals are the best raw materials to produce ethanol. The cost of ethanol production is an essential factor in order to use ethanol for synthetic gasoline production.

The world's documented petroleum deposits (1033 billion barrels) are estimated to be sustained for 39 years, assuming the current consumption rate of petroleum.

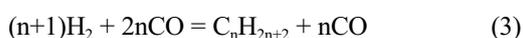
The culmination point for petroleum extraction is expected to happen in 2010 [3]. A similar estimation concerning natural gas deposits and its consumption rate reveals that its global deposits will suffice for the next 63 years. For the time being, the world's hard-coal deposits are estimated to be 514 billion tons (12.1 billion in Poland), and the deposits of brown coal are estimated at 485 billion tons, out of which 13.2 billion tons are in Poland. Though the percentage share of coal used for energy production diminished from 51% in 1910 to 22% in 1997, its global consumption has increased by 312% within this period. This means that during the implementation of technology for the production of synthetic gasoline based on ethanol, or generally speaking based on alcohols, the energy required to run the process will come from hard or brown coal. Plants to produce synthetic gasoline have operated several decades in South Africa under the name of SASOL. The technology is based on the Fisher-Tropsch reaction (F-T). The production of engine fuels by the F-T method from a mixture of gases containing carbon oxide and hydrogen is carried out on the iron and cobalt catalysts, according to the following chemical equations for the chemical reactions:



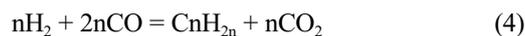
...and



If cobalt catalysts are used, the equations for the chemical reactions are:



...and



The obtained mixture of hydrocarbons is then subjected to the distillation operation and separated into liquid-phase gas, gasoline, jet fuels, diesel oil, and paraffins fractions.

There are several technological differences within respective technologies and, as regards the classical MOBIL method called the MTG process (methanol to gasoline), the gasoline is produced via the methanol synthesis. Such obtained gasoline meets the Eurosuper-95 norm/standard.

The patented processes to produce engine fuels based on the reagent mixture of CO+H₂ are shared by the following consortia: Mobil, BP, Conoco, Rewtech, Sasol-Chevron, Moss gas, Shell, and Syleum. They differ in technological realizations and investment expenditures. The essence of the MOBIL method and other processes based on this idea lies in omitting the direct reaction of CO or CO₂ and H₂ to get synthetic gasoline. Instead of this, the proposal is to synthesize gasoline through methanol or its homologues. The result is such that the equations for the chemical reaction are simplified, because we only deal with the intermediate product.

The key for the synthesis is a catalyst based on ZSM-5 zeolite. As of today, there are known series of more active catalysts when compared with their original ones, and that are also based on a aluminosilicate matrix. However, in the case of the MOBIL synthesis there is a possibility that during the process water will be formed as a by-product. The equation for the synthesis reaction of which the mixture of hydrocarbons is synthetic gasoline can generally be written:



The numerical values of n and x depend on the temperature and pressure under which the process of synthesis is carried out. Water formed during the reaction is undesirable since zeolite catalysts are sensitive to the presence of water.

In the case of using ethanol as a basic new material, the number of reaction cycles is diminished and the yield of synthesis increases. It means that the key factor to get an appropriate mixture of hydrocarbons is the catalyst, which besides its catalytic function in the synthesis process should also bring about isomerization effects of hydrocarbons that enhance the octane number of gasoline and improve the combustion process. Ethanol coupling to higher hydrocarbons is called ETG.

From MTG to ETG

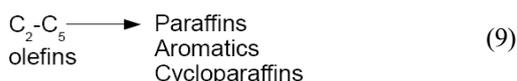
The formation of water during the running of the process causes classical catalysts employed for the alcohol coupling toward gasoline to lose their high activity and selectivity at the H₂O concentration as low as even 100 ppm. Conducting research over several years, the Department of Environmental Chemistry of the Faculty of Chemistry of the University of Maria Curie-Skłodowska in Lublin has worked out a new recipe of a catalyst that resists

large water vapor excesses present in the process, even more than 10 % vol. This means that the couplings process undergoes a substantial simplification, and this results in a significant reduction of the investment to build a plant. The above leads to the relatively lower costs of gasoline production.

The resistance of the catalyst for the water presence has enabled us to realize a gasification process of carbon deposits formed on the catalyst surface that courses to sustain the catalyst activity and selectivity that no longer depends on the time of the running process. Therefore, it has been possible to work out a new process called ETG which, as a consequence, gives the alternative possibility to produce both gasoline and diesel oil that is exactly the same as those obtained from petroleum. Thus, the ETG process opens the alternative source of raw materials in respect to the petroleum import. The equation for the synthesis reaction of a hydrocarbon mixture in the ETG process can be written analogously to that for the MTG process, namely:



Similarly, as for the MTG process, the reaction proceeds through the surface fragment of CH_3OCH_3 (DME), the formation of which is facilitated because the process reactant is the molecule of ethanol and not methanol. Subsequent reactions of ethanol and DME coupling seem to be analogical to the reaction mechanism proposed by Chang et al. [4] for the MTG process, but without the formation of water:



Further coupling of ethanol, however, leads to the formation of water molecules, the amount of which in the ETG process is reasonably smaller than that in the MTG process.

There is also a possibility to run the process but the coupling of earlier formed surface olefins and paraffins with ethyl alcohol, and to be more accurate, with their dehydrogenated surface fragments toward ethers and particularly to ethylene tertbutyl ether (ETBE), and this is consistent with the observed mechanism for the MTG reaction described by Chang [5]. Chen and Reagan suggest that the coupling reaction takes place between DME, alcohol and olefins and leads to the formation of oxygenates of the ether type [6], and this was indirectly confirmed by Chang [7], who modified the Chen and Reagan mechanism and who concluded that:

$$\text{A} = \text{B} \quad (10)$$

$$(\text{A} + \text{B}) = \text{C} \quad (11)$$

$$(\text{A} + \text{C}) = \text{C} \quad (12)$$

$$\text{C} = \text{D} \quad (13)$$

...where: A - alcohol and DME, B - surface CH_2 groups, C - olefins, D - paraffins and aromatics.

In the case of the ETG process mechanism, the following reaction that leads to higher ethers should also be included:

$$(\text{A} + \text{C}) = \text{E} \quad (14)$$

...where: A - alcohols and DME, C - olefins, E - ethers.

The numerical values of n , and x coefficients that are present in equation 6 depend on the temperature and pressure of the process, and also depend on the type of catalyst used. Similarly, as in the case of the ETG synthesis, during running of the process, a good amount of heat is evolved. For that reason, the reactor is divided into two sections and is equipped with a heat exchanger. It is necessary to emphasize that because of the properties of the new generation of the catalysts, the reacting mixture is not subjected to circulation in the reactor with circulated gases, since the coupling process requires one-step only. The above simplifies the technology and, at the same time, it lowers both investment and production costs. It is obvious that the key factor to get the desired composition of a hydrocarbon mixture is using the appropriate catalyst which, besides its function responsible for the synthesis, should also course isomerization effects of hydrocarbons that are crucial for the enhancement of the octane number of gasoline and also improves its combustion process. Analogously to the classical synthesis based on methyl alcohol (MTG) mentioned earlier, the heat effect of the reaction is very high and this fact has to be taken into account at the time of designing the reactor, especially when building large plants. Similar to the MTG classical process, the yield toward engine gasoline produced with the use of zeolite catalysts is equal to at least 85%, calculated in relation to the obtained hydrocarbons. Such obtained gasoline does not contain sulphur or nitrogen, but fortunately contains a significant fraction of ethers.

Catalyst Proposed for ETG Synthesis

Several published papers describe the procedures to obtain catalysts through the cation or anion exchange methods [8, 9]. To obtain the catalyst Ferrierite-Cu, a basic material called ferrierite was used. The general formula for ferrierite is $(\text{Na},\text{K})_2\text{Mg}[\text{OH}/\text{Al}_3\text{Si}_{15}\text{O}_{36}] \cdot 9\text{H}_2\text{O}$. This substance crystallizes in the orthorhombic system. Its hardness is 3-3.5 in Mohr scale and its color is white or yellowish, and having a glassy silken luster. Usually it forms crystals of a needle or hair type. It is also brittle and transparent [10]. The active surface area of the used ferrierite for our research was $34.5 \text{ m}^2/\text{g}$ and was determined with the BET method (adsorption of argon at the temperature of liquid nitrogen). The sample did not contain aluminum except that bound into the net. Copper was introduced to ferrierite with the ion exchange method (using the ammoniacal complex of

Table 3. Physico-chemical properties of studied catalysts based on ferrierite.

Catalyst designation	Cu content, wt. %	Number of moles in the sample		Mole ratio of SiO ₂ /Al ₂ O ₃
		SiO ₂	Al ₂ O ₃	
F-0	0.000	1.13	0.11	10.35
F-1	0.026	1.13	0.11	10.35
F-2	0.052	1.13	0.11	10.35
F-3	0.108	1.13	0.11	10.35
F-4	0.218	1.13	0.11	10.35
F-5	0.446	1.13	0.11	10.35
F-6	0.926	1.13	0.11	10.35

copper), changing the concentration of the impregnate solution (up to 5 wt. %) but without changing the impregnation time. Obtained catalysts at their precursors stage were analyzed with XRF, FT-IR, and then after calcining in air for 2 hrs at 500°C, they were examined with XRD, TPO and TPD methods. Then the rates of the reaction and the selectivity of the process were determined by means of a gradientless reactor described elsewhere [11]. The rate of reaction (TOF – Turnover Frequency) was calculated from equation (15), which is valid for the gradientless reactor [11, 12]:

$$r = [(C_{we} - C_{wy})/C_{we}] \cdot F / [(V_{Ar}/1000)W] \quad (15)$$

...where:

r – the reaction rate (TOF) (s⁻¹),

F – the volumetric flow of C₂H₅OH through the reactor (mole/s),

C_{we} and C_{wy} – the inlet and outlet concentrations of C₂H₅OH, respectively (mole ratios),

W – the mass of catalyst samples used in the experiment (g),

V_{Ar} – the volume of argon adsorbed by the sample at Θ (degree of surface coverage) = 1, (μ mole/g, it was also assumed that it was equivalent to the number of moles of active catalyst in the process).

The process selectivity toward appropriate products was computed from equation (16):

$$S = r_p / \sum r_i \quad (16)$$

...where:

S – selectivity (arbitrary units),

r_p – the reaction rate toward the appropriate product (TOF) (s⁻¹),

$\sum r_i$ – the sum of the reaction rates toward all analyzed products (TOF) (s⁻¹).

The changes in the reaction rates at the temperature of 653 K, and the constant conversion rate (X) equaled to 0.05 (5%) were studied. The physico-chemical properties of used catalysts based on ferrierite are presented in Table 3.

It was assumed that in the kinetic tests the important feature of the catalyst activity would be the reaction rate TOF

toward the formation of ETBE as the most desirable product, and also toward the formation of hydrocarbon isomerization containing more than 5 carbon atoms in a molecule. Fig. 2 shows changes in the reaction selectivity of studied ferrierite catalysts vs. the function of the Cu concentration in the catalysts, at a temperature of 653 K and at the degree of the C₂H₅OH conversion equal to 5% ($X=0.05$).

The obtained results prove that there exists a certain optimum in the Cu concentration in catalysts, and this optimum lies in the range of 0.2-0.4 wt.% of copper. The reaction selectivity toward ETBE (ethyl tert-butyl ether) is, however, the highest at somewhat greater concentrations of the metal (0.5 wt.% Cu). At copper concentrations greater than 0.5 wt.%, the selectivity increases but, unfortunately, undesirable products like CH₄, CO₂ and CO are formed. Raw ferrierite (the catalyst designated as F-0) exhibits a low selectivity toward desired reaction products.

Table 4 presents values of the apparent activation energy for examined catalysts.

The introduction of Cu to the catalysts reasonably changes the values of the apparent activation energy that

Table 4. Activation energy for ferrierite catalysts supported with Cu.

Catalyst designation	E_{total} , [kJ/mol]	ΔE , [kJ/mol]
F-0	128.3	±21.2
F-1	154.5	± 4.8
F-2	176.5	±11.1
F-3	275.7	±25.2
F-4	309.6	±41.2
F-5	312.7	±46.1
F-6	298.5	±20.3

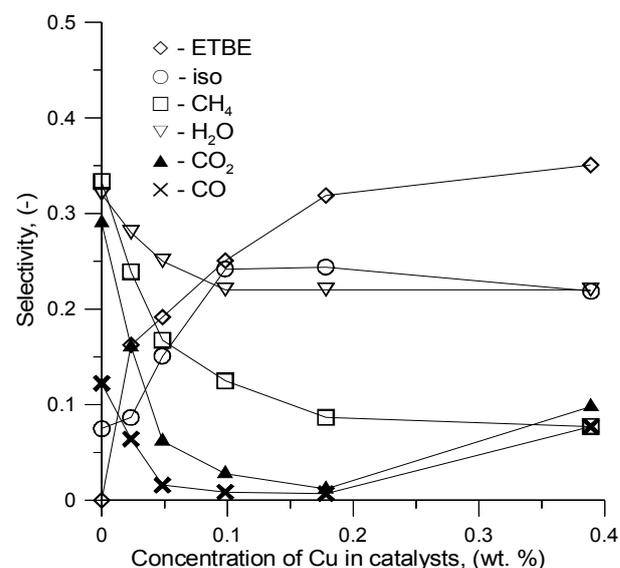


Fig. 2. Changes in process selectivity of the ethanol coupling reaction as a function of the Cu concentration in catalysts, at 653 K and at the conversion degree of 5% ($X=0.05$).

Table 5. Variations in selectivity of the ethanol coupling reaction at X = 80% and T = 653 K for only oxygenates and hydrocarbon products [P-384999].

Activation energy [kJ/mol]	Reaction selectivity [%]				
	ETBE and other ethers	iso-hydrocarbons	SNG*	CO, CO ₂	Other hydrocarbon fractions
270.1	48.9	32.8	10.8	2.2	5.3

* SNG – Substitute Natural Gas.

Table 6. Changes in selectivity of the ethanol coupling reaction at X = 80% and T = 693 K for only oxygenates and hydrocarbon products [P-384999].

Activation energy [kJ/mol]	Reaction selectivity [%]					
	ETBE and other ethers	iso-hydrocarbons	Paraffins	SNG	CO, CO ₂	Other hydrocarbon fractions
287.9	37.7	22.6	14.3	2.2	5.3	25.4

Table 7. Changes in selectivity for the butanol coupling reaction at X = 80% and T = 653 K for only oxygenates and hydrocarbon product [P-384999].

Activation energy [kJ/mol]	Reaction selectivity [%]								
	ETBE and other ethers	Iso-hydrocarbons	Paraffins	Aromatic fraction	Glicol	Durene	SNG	CO, CO ₂	Other hydrocarbon fractions
291.3	17.8	20.5	16.9	9.8	6.3	2.6	3.4	2.5	20.2

rapidly increases from 128.3 kJ/mol to 312.7 kJ/mol for the F-5 catalyst. Conducted studies reveal that the most favorable catalyst obtained by a preparation method described by [4, 5] should possess the following physico-chemical parameters:

1. S_{total} – total surface area of 35 m²/g,
2. Cu concentration in the catalyst – 0.2-0.5 wt.%,
3. absence of three-dimensional Cu crystallites,
4. ratio of SiO₂/Al₂O₃ = about 10,
5. apparent activation energy of reaction (E), about 300 kJ/mol,
6. selectivity toward (calculated for only oxygenates and hydrocarbon products):
 - a) ETBE – about 45% ,
 - b) C₅₊ isomers – about 30%,
 - c) CO₂ and CO – less than 12%.

ETG Process, Reaction Products for a System of Higher Conversions and Higher Pressures

The essence of the novel realization for the ETG process leading to the hydrocarbon synthesis employs a new generation of catalysts that are resistant to reasonable excesses of water vapor during the process [P-384999]. Using such catalysts eliminates a coking process at a deep dewatering operation of circulated gases, and also eliminates a need to dewater the inlet reagents used for the synthesis. It also enables the running of the process even when the reacting mixture contains up to 10 vol. % of H₂O.

The substitution of methanol with its homologue C₂H₅OH, results in the lowering of the pressure required to conduct the process from 300 to 30 bars.

Table 5 shows the changes in process selectivity for a temperature of 653 K and pressure of 10 bar for ethanol as a substrate. At this temperature any durene (C₆H₂(CH₃)₄) fraction occurs. Table 5 also shows the variations in the process selectivity at the constant conversion ratio of X=80% [P-384999].

For a temperature of 693 K and pressure of 14.4 bar the changes in the reaction selectivity are observed and these changes are presented in Table 6 [P-384999].

The substitution of ethanol with butyl alcohol widens a spectrum of raw materials, and it also results in the lowering of the operation pressure. For example, at a temperature of 653 K, X=80% and under a pressure of 4 bars, we get the following variations in the process selectivity (Table 7) [P-384999].

From the obtained products, and after their separation by the fractionating distillation, they can serve as components to prepare a commercial mixture, this is gasoline characterized by a small octane number, or diesel oil. Based on the characteristic data related to MTG processes it is possible to compute a rough-estimate cost for the ETG plant processing 50,000-5,000,000 tons of ethanol as a raw material [13]. For a plant processing 500,000 tons of ethanol per year, the investment cost to build the plant will be of the order of 520 mln PLN, and the simplified scheme diagram of such a plant is shown in Fig. 3 [14]. The obtained mixture of hydrocarbons can be used to prepare

Table 8. Comparison between the compositions of synthetic gasoline and Euro95 [14].

Sulphur content	LOB	Aromatics	Benzene	Toluene	Xylene	TBA*	DIPE**	ETBE	TAME***	MTBE****	Total oxygen	Olefins	Saturated organic compounds
		vol.%											
0.01	95.8	37.09	2.32	9.86	9.42	0.21	-	0.27	0.48	5.63	1.18	9.71	46.6
-	107.9	16.32	-	-	-	-	-	22.76	-	-	3.28	8.57	52.35

TBA* – tert-butanol, DIPE** – diisopropyl ether, TAME*** – tert-amyl methyl ether, MTBE**** – methyl tert-butylene ether.

required blends of diesel oil of the desired cetane number, especially that of the order of 56.

It is also obvious that in order to give the exact expenditures it is necessary to work out a precise project/report that should include all costs, especially the costs of raw materials and licences. These charges may cause the total cost of the investment to vary by $\pm 10\%$ [13].

Table 8, compares the compositions of synthetic gasoline obtained from ethanol with a classical commercial petrochemical product, i.e. Euro95 gasoline [14].

As seen in Table 8, the synthetically obtained product does not contain both benzene and sulphur compounds, and it means that this biofuel fulfils several environmental requirements more than for Euro95 [14].

Conclusions

Based on the presented results of our studies and on a brief evaluation of the market, it can be stated that the application of alcohol as a raw material for the production of biofuels may be highly challenging. The preparation recipe for obtaining the new, highly-active and selective catalyst enables us to conduct this process. It is also neces-

sary to emphasize that in the case of Poland, fuel obtained this way will replace adequate amounts of imported petroleum.

The raw material (ethanol) can come from the domestic sources or can be cheaply imported from Ukraine and Brazil [13]. Taking into account two facts, namely:

- (1) that the sources from which bioethanol is produced belongs to a class of renewables, and
- (2) the composition mixture of hydrocarbons obtained in the coupling reaction of ethanol is quite different from the composition of petroleum; therefore, we can conclude that the biofuel obtained via ethanol is a fuel of the second generation.

Actually, in this case, similarly to the case of MTG, there is no need to adapt existing gasoline engines to the new fuel mixture, and this makes the technology attractive to the fuel market.

It is also necessary to point out that the ETG fuel is much more pure than the classical one, and this is the result of using ethanol as a substratum. Therefore, this type of fuel is much more friendly for the natural environment and also for the combustion engine itself. This note concerns especially diesel oil obtained according to the described ETG process [14].

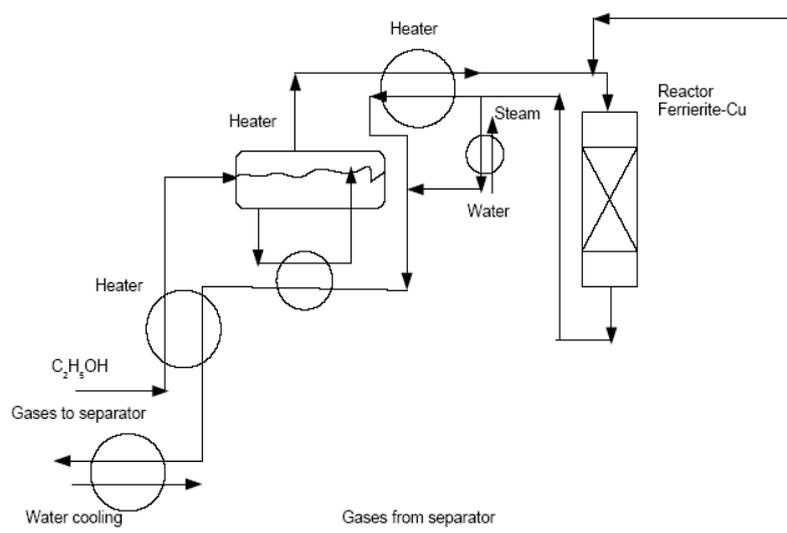


Fig. 3. The scheme diagram of ETG process [14].

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