Mathematical modeling of biodiesel production process

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Abstract
The article presents the results of theoretical and experimental studies of the processes of transesterification of vegetable oils with methyl alcohol, the material balance is determined, and the molecular weights of the components involved in the process of transesterification as input and output products are established. The influence of temperature and changes in the critical mixing rates on the acceleration of the process of transesterification of vegetable oils with methyl alcohol, the results of an experimental study of the processes of biodiesel production by mobile equipment.

Keywords: biodiesel, transesterification processes, biodiesel production, diesel internal combustion engines, reduction of emissions from diesel engines

INTRODUCTION

The intensive development of industry and transport infrastructure in the modern megacities of the country has led to a deterioration of environmental tensions. One of the most perspective solutions to this problem is the use of alternative fuels for diesel engines.

Due to fuel crises and the adoption in developed countries of strict standards on exhaust gas toxicity, in recent years, studies have been conducted and new technologies have been introduced for the production of fuel for use in carburetor and diesel engines from renewable energy sources.(Ragulin et al. 2019)

Biodiesel was first proposed by Englishmen Duffy and Patrick. Only 40 years later, the German scientist Rudolf Diesel invented an engine running on peanut oil. Diesel fuel at that time was cheaper, so it became for many years the main fuel for diesel engines (Fuchs et al. 1992, Mitin 2007).

Transport biofuels exist mainly as ethanol and biodiesel. In 2014, ethanol accounted for 74% of the transport biofuel market, biodiesel - 23% (mainly in the form of methyl esters of fatty acids), hydrogenated vegetable oil (HVO) – 3%. These fuels are produced from food raw materials. Ethanol is obtained from sugar cane (61%) and grain (39%). The main raw materials for the biodiesel production are soybeans and rapeseed. Attempts to commercialize liquid biofuels from sources that do not compete with food production so far failed to produce statistically significant market results.

The Methodology and Results of Research

The rate of the transesterification process using alkaline catalysts largely depends on the concentration of alkalis in the mixture, the temperature of the process and the pressure in the reaction zone. The scheme of the process of transesterification of vegetable oils with methyl alcohol is shown in **Fig. 1**.
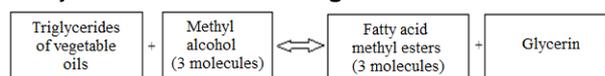

Fig. 1. The scheme of the process of transesterification of vegetable oils with methyl alcohol

An increase in the concentration of the alkaline catalyst accelerates the process of transesterification, but when using highly concentrated alcoholic solutions of alkalis, simultaneously with transesterification, a significant saponification of fats can take place. A series of experimental studies aimed at deepening knowledge in two directions has been carried out in ITTP NAS of Ukraine: the transesterification of vegetable oils with ethyl alcohol and the preparation of mixtures of vegetable oils and alcohols (Grabov and Shmatok 2008, 2009).

To conduct experimental studies in the direction of transesterification of vegetable oils with methyl and ethyl alcohols, it was necessary to determine the material balance of the process. For this purpose, the molecular
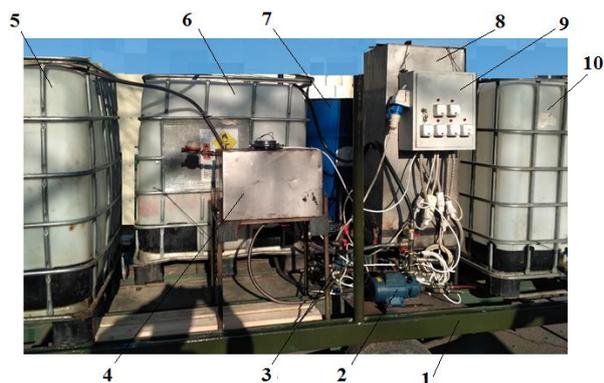
1 - mobile equipment casing; 2-3 - pumps; 4 - methoxide unit - preparation and dosing of methoxide in automatic mode; 5 - methanol recovery complex from glycerol; 6 - gravity separation unit - separation of biodiesel and glycerol; 7 - oil unit - dosing of oil in automatic mode, 8 - biodiesel synthesis reactor, 9 - control panel, 10 - unit for finished glycerin, 11 - filter unit - filtration of finished biodiesel

Fig. 2. General view of mobile equipment

weights of the components involved in the process of transesterification as input and output products were determined.

Acceleration of transesterification is possible by increasing the temperature and pressure, but the increase in technological parameters leads to a complication of technology and equipment, which can have a negative impact on the cost of fuel produced. Therefore, the selection of rational technological parameters of production is an important task.

In the proposed devices (Aldabergenov et al. 2017, 2018, Kumaran et al. 2013, Palash et al. 2013, Roy et al. 2013) for the production of biodiesel from fat-containing waste, as well as to accelerate the transesterification reaction, a patented new method is proposed.

As a result of theoretical and experimental studies, the optimal parameters of mobile equipment, working processes of biodiesel production and its application in the vehicle were established.

Mixing the normalized fluxes of the source components created by the rotor of the centrifugal pump-activator consisting of a rotor, the walls of the pump casing, between which the area of hydrodynamic mixing of the components of the inlet of the pump-activator, pipelines of oil flow, flow of alcohol with the catalyst, outlet. In this case, the input flow of the source components in the supply pipelines is formed.

In the method of activation of the transesterification reaction in the preparation of fatty acid esters, including the reaction of vegetable oil with alcohol in the presence of a catalyst, hydrodynamic mixing is used - activation due to the energy of the rotating rotor of a standard centrifugal pump, at a rotor rotation rate of 1200-2600 rpm and a temperature of 57°C.

The method is protected by a positive decision No. 871 of 10.07.2017 on the issuance of a patent of the Republic of Kazakhstan for the invention on application

No. 2016/0983,1 "Method for activating the transesterification reaction in the preparation of fatty acid ether."

Loading of components into the reactor, mixing, unloading of homogeneous mixture for further gravitational separation of finished products obtained at the end of the chemical reaction is carried out by one mechanism of a centrifugal pump using one input channel.

Using the proposed method, mobile equipment was created (**Fig. 2**).

The equipment consists of a two-axle triller (trolley) mounted on the casing, which provides full mobility of the equipment, pumps, a methoxide unit – for the preparation and dosing of methoxide in automatic mode, a methanol recovery complex from glycerol, a gravity separation unit - separation of biodiesel and glycerol, an oil unit - dosing of oil in automatic mode, a bio-diesel synthesis reactor, a control panel, a unit for finished glycerol, a filter unit - filtration of finished biodiesel.

The process of biodiesel production is considered, which is a methanolysis reaction (interaction of triglycerides of vegetable oil with methanol) occurring in an apparatus with a working volume $V(m^3)$ in the presence of alkali (as a catalyst) when the initial components are heated (to accelerate the process).

After building the physico-chemical model required for specifying the main substances involved in the biodiesel production process will proceed to describe its mathematical model including balance equation of the apparatus flows, the equation balances for each of the substances involved in the reactions and the equation of heat balance. The concentration of substances at the inlet of the apparatus c_{iin} is expressed

$$c_{iin} = \frac{G_i}{v_{in}} \qquad (1)$$

where G_i – mass flows of substances involved in the reactions at the inlet of the apparatus, kg/s; v_{in} - total volumetric flow rate of liquids entering the apparatus, m^3/s.

The following source components are loaded into the apparatus:

- vegetable oil in the amount of 100 kg (109.3 l);

- including the components (**Table 1**), considering that the source oil - mono glycerides are absent.

Table 1. Percentage ratio of fatty acids, which are included in the composition of vegetable oils (Kuliev et al. 1999)

№	Fatty acids included in triglycerides of vegetable oils	Chemical formula of acid	Molecular weight, g/mol	Composition of acids, % mass flows of substances		
				Rapeseed oil	Sunflower oil	Soybean oil
1.	Palmitic	$C_{16} C_{32} O_2$	256.42	4.35	8.92	14.22
2.	Stearic	$C_{16} H_{30} O_2$	284.47	2.62	4.16	4.54
3.	Oleic	$C_{16} H_{34} O_2$	282.45	25.75	22.63	25.63
4.	Linoleic	$C_{18} H_{32} O_2$	280.44	23.77	61.46	46.42
5.	Linolenic	$C_{18} H_{30} O_2$	278.42	29.25	1.12	7.21
6.	Arachidic	$C_{20} H_{40} O_2$	312.52	0.59	0.11	0.38
7.	Eicosenoic	$C_{20} H_{38} O_2$	310.50	7.91	0.67	0.56
8.	Eicosadienoic	$C_{20} H_{30} O_2$	308.48	0.55	0.16	0.21
9.	Erucic	$C_{22} H_{42} O_2$	338.56	4.26	-	-
10.	Nervonic	$C_{24} H_{40} O_2$	366.61	0.95	-	0.14
11.	Other acids		≈ 300	-	0.77	0.69

Table 2. Mass composition of source components

№	Component	Mass, in (kg/l)	The percentage ratio of the total (%)	Note
1	Fat-containing waste	94,7/100	84,3	
2	Ethyl alcohol	14,2/18	15,1	
3	Catalyst	0,745	0,60	
	Total mass	118,745	100	

Table 3. Concentration of source components (kg/l)

№	Component	Concentration c_{iin}, kg/l	Note
1	Fat-containing waste	0.744	
2	Ethyl alcohol	0.143	
3	Catalyst	0.06	

Table 4. Concentration of source components (mol/l)

№	Component	Concentration c_{iin}, mol/l	Note
1	Fat-containing waste	0.843	
2	Ethyl alcohol	0.151	
3	Catalyst	0.06	

- alcohol catalyst solution in the amount of 14,945 kg (18.7 l);

- the total volumetric flow rate of liquids entering the apparatus is 118.745 liters.

For the substances involved in the reaction, the concentration of the source components in kg/l is shown in **Table 3** and the concentration of the source components in mol/l in **Table 4**.

To obtain the equation of material balance by flows, we present the mass flow rate of the liquid entering the apparatus in the form of production $v_{in} \cdot p_{in}$ here p_{in} is the average density of the flows of substances entering the apparatus, equal to 0,882 kg/l.

Then the mass flow rate of the liquid entering the apparatus:

$$v_{in} \cdot p_{in} = 118{,}745 * 0{,}882 = 104{,}733 \text{ (kg)}.$$

The total flow of substances at the outlet of the apparatus is recorded as vp. Here p is the density of the liquid in the apparatus. For the process under study, the

synthesis results in: methyl esters of higher unsaturated aliphatic acids $E_1–E_2$; a mixture of glycerol and potassium glycerate; methyl alcohol that has not reacted.

The total flow of substances at the outlet of the apparatus is recorded in the form: vp =118,745*0,891=105,802 (kg). In this case, the sum of the input and output flows is equal to the rate of change in the mass of the liquid in the dVp/dt apparatus, or

$$\frac{d(Vp)}{d\tau} = v_{in} p_{in} - vp \qquad (2)$$

where τ - time, s.

In our case for a stationary process we get:

$$\frac{d(Vp)}{d\tau} = v_{in} p_{in} - vp = (118.745 * 0.882) - (118.745 * 0.886) = 0.003.$$

Equation (2) is the equation of the material balance for flows in the apparatus. In addition to the derived equation, it is necessary to obtain the equations of material balance for individual substances involved in the reaction. Write down the rate of change in the mass of the i-th substance $d(Vc_i)/d\tau$, located in the apparatus, in the form:

$$\frac{d(Vc_i)}{d\tau} = v_{in} c_{iin} - vc_i + VR \qquad (3)$$

where $v_{in} c_{iin}$ – the flow of substance at the inlet to the apparatus or the number of substances entering the apparatus per unit time, kg/s; vc_i - the flow of substance at the outlet of the apparatus, kg/s; R_i - the total rate of formation (consumption) of the i-th substance, kg/m³ s.

Since the value of the quantity R_i consists of individual rates of reactions for this substance, it is represented as:

$$R_i = \sum_{j=1}^{q} rij \qquad (4)$$

where r_{ij} is the rate of the j-th reaction on the i-th substance,

$$rj = \frac{r_{1j}}{M_1 a_{1j}} = \frac{r_{2j}}{M_2 a_{2j}} = \dots idem \qquad (5)$$

where M_i is the molecular weight of the i-th substance; $\alpha_1 \dots \alpha_n$ - stoichiometric coefficients equal to the number of molecules of the source substances; $\alpha_{n+1} \dots \alpha_m$ - stoichiometric coefficients equal to the number of molecules of the reaction products;

Analysis of equation (4) and (5), shows that

$$r_{ij} = M_i \alpha_{ij} r_i = M_i \alpha_{ij} k_j c_1^{n_{1j}} c_2^{n_{2j}} \dots c_m^{n_{mj}} \qquad (6)$$

Then the expression for R^1 taking into account (6) will take the form

$$R_1 = M_i \sum_{j=1}^{q} \alpha_{ij} r_j = M_i \sum_{j=1}^{q} \alpha_{ij} k_j \prod_{s=1}^{m} c_s^{n_{sj}}. \qquad (7)$$

When substituting the obtained expression (7) into equation (3), the final equation of the material balance for the individual substances involved in the synthesis reaction is obtained,

Table 5. Total consumption rates of components (source mixture)

№	Component	Consumption rates, R_i, mol/l s	Note
1	Fat-containing waste	$1.0 \cdot 10^{-4}$	
2	Ethyl alcohol	$4.26 \cdot 10^{-6}$	
3	Catalyst	$3.7 \cdot 10^{-7}$	

Table 6. Concentration of reaction products (kg/l)

№	Component	Symbol	Concentration c_{iin}, kg/l	Note
1	Fat-containing waste	E_1	0.744	
2	Ethyl alcohol	E_2	0.143	
3	Catalyst	E_3	0.06	
4	Glycerin	G	0.066	
5	Methyl alcohol	MA	0.134	
6	Di- and monoglycerides	D+M	0.043	

Table 7. Concentration of source components (mol/l)

№	Component	Symbol	Concentration c_{iin}, mol/l	Note
1	Fat-containing waste	E_1	1.643	
2	Ethyl alcohol	E_2	0.551	
3	Catalyst	E_3	0.106	
4	Glycerin	G	0.71	
5	Methyl alcohol	MA	4.13	
6	Di- and monoglycerides	D+M	0.093	

Table 8. Total formation rates of final products of reaction

№	Component	Symbol	R_i, mol/(l-s)	Note
1	Fat-containing waste	E_1	$2.36 \cdot 10^{-4}$	
2	Ethyl alcohol	E_2	$4.92 \cdot 10^{-5}$	
3	Catalyst	E_3	$1.51 \cdot 10^{-4}$	
4	Glycerin	G	$5.82 \cdot 10^{-5}$	
5	Methyl alcohol	MA	$9.96 \cdot 10^{-5}$	
6	Di- and monoglycerides	D+M	$1.32 \cdot 10^{-5}$	

$$\frac{d(Vc_i)}{d\tau} = v_{\text{вх}}c_{i\text{вв}} - vc_i + VM_i \sum_{j=1}^{q} \alpha_{ij}k_j \prod_{s=1}^{m} c_s^{n_{sj}} \qquad (8)$$

The studies of the kinetics of the studied reactions allowed to establish (**Table 5**) the total consumption rates of the source components R_i. The concentrations of these components at the inlet were calculated earlier, at the outlet - are equal to zero. At the end of the reaction, the concentration values (expressed in kg/l, presented in **Table 6**, and in mol/l - in **Table 7**) of methyl esters of higher and unsaturated aliphatic acids (E_1-E_3), glycerin (G), unreacted methyl alcohol (MA), di - and mono–glycerides are absent in the reaction mass.

The concentration of methyl esters of higher unsaturated aliphatic acids (E_1-E_3), glycerin (G), di- and monoglycerides in the source mixture (at the reactor inlet) is zero.

The total formation rates of R_j final products are presented in **Table 8**.

Based on certain molecular weights of the input and output products of the transesterification reaction and the scheme of the transesterification process of vegetable oils with methyl and ethyl alcohols (**Table 9**), we calculate the material balance of the process of transesterification of vegetable oils (rapeseed, sunflower and soybean) with methyl and ethyl alcohols, respectively.

The mass of the resulting esters is determined by the formula:

Table 9. Molecular weights of the components involved in the transesterification process as input and output products

Substance	Molecular weight, g/mol
Rapeseed oil RO	894.3
Sunflower oil SrO	876.0
Soybean oil SnO	872.5
Methyl alcohol	32.0
Ethyl alcohol	46.1
Methyl ester RO	299.5
Methyl ester SrO	293.4
Methyl ester SnO	292.2
Ethyl ether RO	313.5
Ethyl ether SrO	307.4
Ethyl ether SnO	306.2
Glycerin	92.1

Table 10. Calculation of material balance of transesterification of 1 kg of vegetable oils with methyl alcohol

Oils / Alcohol	Rapeseed oil	Sunflower oil	Soybean oil	Fat-containing waste
Methanol	mal = 0.1075 kg mes = 1.0045 kg mgl = 0.1030 kg	mal = 0.1098 kg mes = 1.0047 kg mgl = 0.1051 kg	mal = 0.1102 kg mes = 1.0047 kg mgl = 0.1055 kg	mal = 0.112 kg mes = 1.027 kg mgl = 0.095 kg

$$m_{es} = 3 \cdot m_o \cdot M_{es}/M_o \qquad (9)$$

where m_{es} – mass of esters; m_o – mass of oil; M_{es} – average molecular weight of esters; M_o – molecular weight of oil.

Mass of glycerin produced:

$$m_{gl} = m_o \cdot M_{gl}/M_o \qquad (10)$$

where m_{gl} – mass of glycerin; m_o – mass of oil; M_{gl} – molecular weight of glycerin; M_o – molecular weight of oil.

The mass of alcohol necessary:

$$m_{al} = 3 \cdot m_o \cdot M_{al}/M_o \qquad (11)$$

where m_{al} – mass of alcohol; m_o – mass of oil; M_{al} – molecular weight of alcohol; M_o – molecular weight of oil.

According to the formulas (9), (10) we determine the mass of esters and glycerin, which are formed in the process of transesterification of oils with methyl and ethyl alcohols. Also, according to the formula (11), we determine the mass of the required alcohol (methyl or ethyl). Calculations were carried out for 1 kg of oils. The results of the calculations are presented in **Table 10**.

The calculations show that the material balance of transesterification with the use of different oils is practically the same with the use of methyl alcohol. The maximum difference in the amount of alcohol required for the transesterification of different oils is 2,7% when using methyl alcohol. Thus, it can be concluded that the type of oil practically does not affect the material balance of the transesterification process in the case of methyl alcohol.

To determine the rational parameters of obtaining biodiesel fuel by transesterification of vegetable oils with methyl alcohol, experiments were carried out on an experimental stand for the study of heat and mass transfer processes of biofuel preparation (**Fig. 3**).
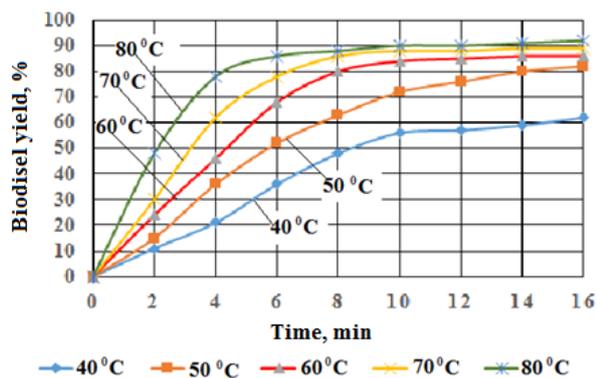
Fig. 3. Dependence of biodiesel yield on temperature at KOH = 1.2%

Fig. 4. Dependences of biodiesel yield on changes in the critical mixing rate V_{cr} at a content of KOH = 1,3% and a temperature of 60°C

The excess alcohol coefficient K is defined as the ratio of the mass of alcohol that is used to the mass of alcohol theoretically necessary for complete transesterification:

$$\text{K} = {m_{al}^{a}}\big/{m_{al}^{t}} \qquad (11)$$

where K – coefficient of excess alcohol; m_{al}^{a} – actually used mass of alcohol; m_{al}^{t} –theoretically calculated mass of alcohol.

The results of experimental studies using methyl alcohol are presented in the form of graphical dependences of biodiesel yield on temperature at a content of KOH = 1.2% (**Fig. 3**). During the experimental studies, the influence of individual parameters of the process on the rate and completeness of biodiesel yield was determined.

From the above dependences it can be seen that the rate and completeness of the transesterification process increases with an increase in the temperature parameters of the reaction and with an increase in the amount of alcohol introduced.

At insufficiently high temperatures (30-40°C), the process of transesterification is extremely slow and may not take place completely even with a sufficiently long reaction time. This is due to the fact that the process of transesterification is reversible.

It is experimentally determined that the temperature of 50-80°C is rational for complete transesterification. At such temperature parameters, the process of transesterification is complete in a fairly short period of time and at the same time does not require significant energy costs.

Studies conducted with ethanol (alcohol content 96%, in-dy 4%) have shown that at process temperatures of 40-100 °C, the process of transesterification is extremely slow and is not completed by the production of esters.

The moisture content in the reaction mixture of oil, alcohol and alkaline catalyst leads to rapid saponification of fats and makes it almost impossible to go through the transesterification process.
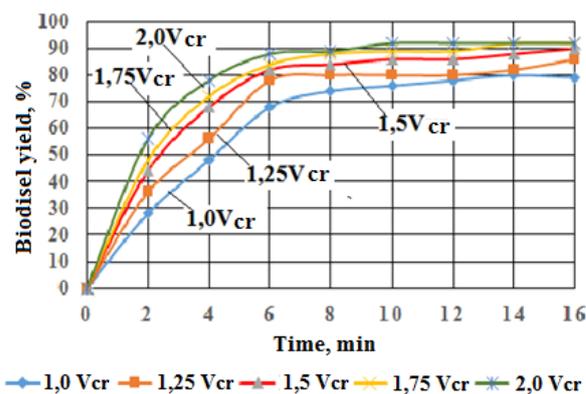
Researches of process of transesterification of vegetable oils of IVA with the content of ethyl alcohol of 99,8% are also carried out. During the experiments, the influence of individual parameters of the process accelerator on the rate and completeness of the formation of ethyl ethers was determined.

In our proposed method, the activation of the transesterification reaction with obtaining esters of fatty acids, comprising the reaction of vegetable oil with alcohol in presence of catalyst, use of hydrodynamic mixing – activation due to the energy of the rotating rotor standard centrifugal pump, when the rotor rate 1200-2600 rpm and temperature above 57°C through the mechanism of the accelerator mechanism that allows to create up to a twofold increase in the critical rate mixing of the treated medium (**Fig. 4**).

From the given dependences of biodiesel yield on changes in the critical mixing rate V_{cr} at a content of KOH = 1.3% and a temperature of 60°C, it can be seen that increasing the critical mixing rate up to 2 times dramatically reduces the time of biodiesel yield, and the completeness of the transesterification process increases.

At low critical rates (1.0 V_{cr} to 1.5 V_{cr}) and at a temperature (40-60°C), the transesterification process is extremely slow and may not take place completely even with a sufficiently long reaction time.

It is experimentally determined that critical rates (1.75 V_{cr} to 2.0 V_{cr}) and temperature (60-80°C) are rational for complete transesterification. With such parameters of the critical mixing rate and temperature, the process of transesterification takes place completely in a fairly short period of time and at the same time does not require significant energy costs.

Experimental studies were carried out on a laboratory sample of mobile equipment for the production of biodiesel from fat-containing waste with an assessment of functional, energy and operational and technological indicators.

When the equipment is operated, approximately 970 kg (1100 l) of biodiesel and 160 kg of primary glycerin are obtained from one ton of waste vegetable oils and 250 (120) kg of alcohol (in the presence of 10-15 kg of NaOH catalyst). Consumption of source components depends on their quality.

The productivity of mobile equipment (for 10 loads per day), biodiesel 2...4 t/day and glycerin 0.2-0.4 t/day, the total power consumption of 36.05 kW, while the energy efficiency of the reactor is 0.85 was determined experimentally.

Quality of the received production (biodiesel fuel) corresponds to the established requirements of ST 176-1907-20, LLP-02-2013 is confirmed by the passport No. 1138, laboratory of ChTO.

proposed laboratory sample of mobile equipment for the production of biodiesel from fat-containing waste have been established. The completeness of the transesterification reaction is achieved at the heating temperature of the mixture 50-80°C, and at the critical mixing rate (1.75 V_{cr} to 2.0 V_{cr}), as well as providing optimal pump performance with minimal power consumption and reactor performance.

The results of the analysis of the quality of mobile equipment produced with the help of a laboratory sample - biodiesel fuel from fat-containing waste - fully comply with the established requirements of ST 176-1907-20-LLP-02-2013 - EN14214 "European standard for biodiesel" and respectively ASTM 6751 "American standard for biodiesel" and EN14214 "European standard for biodiesel."

CONCLUSION

Taking into account the results of theoretical and experimental studies, the optimal parameters of the

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