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APPLICATION OF BY-PRODUCTS OF ETHANOL PRODUCTION IN THE FUEL INDUSTRY

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This article analyzes two ways of solving environmental problems in the fuel industry, which arose due to the need to stabilize the concentration of greenhouse gases in the atmosphere. According to the first direction, the ways of using various non-traditional types of raw materials and waste containing sugars for the purpose of producing environmentally friendly types of fuel are considered. According to the second direction, the possibilities of integrated use of ethanol and all by-products formed during its production are analyzed to improve ecology in the fuel industry. We have established the possibility of using the by-products of ethanol production, fuel oil and fuel alcohol, to improve the operational properties of motor fuels used in carburetor and diesel engines. It has been stated that the additives of fuel alcohol and fuel oil increase the octane number of gasoline, improve its fractional composition, as a result of which the completeness of fuel combustion and the uniformity of its supply to the engine cylinders increase. The optimal concentrations of alcohol additives to gasoline are as follows: 5 vol.% and 10 vol.% for fuel alcohols and fuel oil, respectively. It has been established that the addition of fuel oil to diesel fuels increases their cetane number, this causes the mixture to ignite faster and the diesel engine to start. Moreover, with the addition of fuel oil, the temperature of the beginning of boiling decreases which facilitates the starting properties of the fuel, and the viscosity of the fuel mixture decreases which improves the completeness of mixing and transportation in the engine system. The optimal concentration of fuel oil additive to diesel fuel is 10 vol.%.

Keywords: fuel oil, fuel alcohol, octane number, gasoline, cetane number, diesel fuel.

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Introduction

With the entry into force of the UN Framework Convention on Climate Change, the need to stabilize the concentration of greenhouse gases in the atmosphere was recognized. Therefore, many different regulations have been introduced to reduce greenhouse gas emissions worldwide. Industries that generate significant greenhouse gas emissions have to develop solutions to meet the new limit values. In the fuel industry, this has led to the use of renewable and environmentally friendly fuels based on biologically derived raw materials, such as bioethanol and biodiesel. It is also equally important to dispose of all products produced in alcohol production for the purpose of their ecological use.

Two main directions can be identified in solving these environmental problems related to the fuel industry. The first direction is the use of virgin resources and various non-traditional types of raw materials and waste containing sugars for the purpose of producing ecological biofuel. Switching to sustainable biofuels provides environmental benefits such as reduced carbon emissions and harmful waste. The second direction is the complex use of ethanol and all by-products that are formed during its production, for their use in the fuel industry.

According to the first direction, the method of utilization of household food waste, which contains a significant amount of sugars (both soluble and insoluble) for the production of ethanol, is known. In such a process, a separate liquefaction (saccharification) step was included, which quickly reduced the viscosity of the high-solids substrate, which resulted in better mixing of the fermenting microorganisms. The dry residue after saccharification was subjected to preliminary hydrothermal treatment and used as raw material for further fermentation with the formation of ethanol [1,2].

For the production of ethanol, the raw material can be waste paper, which contains a lot of cellulose and does not require energy-intensive thermophysical pretreatment. Disposal of old waste paper (for example, from landfills) by incineration is considered environmentally harmful due to the formation of greenhouse gases and polluting leachate. In this study, an efficient process of enzymatic saccharification of waste paper suspension with its conversion to ethanol was developed. In addition, the separated distillers' residue in this process was a source of methane formation [3].

Lignocellulosic biomass (for example, straw, forestry waste, and seaweed), sewage sludge, and biogenic fractions of mixed municipal waste are valuable raw materials for biofuel production. Based on such raw materials, there are many options for the synthesis of methanol, ethanol, propanols, and butanols. Such alcohols, created on a biological basis and renewable energy sources, open up new opportunities for fuel production and enable a flexible choice adapted to locally available raw materials, as well as the possibility of integrating biofuel production into industrial processes [4]. Such processes include biomass and waste gasification, as well as carbon capture and utilization to obtain synthesis gas, fermentation of sugars from lignocellulosic raw materials, and new, less developed processes, such as synthesis gas fermentation, glycerol conversion, and biogas reforming [4].

Various technologies for the production of fuel ethanol from raw materials containing sucrose (mainly sugar cane), starchy materials, and lignocellulosic biomass are known [5], as well as the main trends of research on their improvement. The complexity of biomass processing is recognized thanks to the analysis of the various stages of conversion of the lignocellulosic complex into fermentable sugar. The paper presents comparative indicators for the three main types of studied raw materials in the production of fuel ethanol. However, it should be noted that the logistics of ensuring a competitive year-round supply of biomass raw materials to a commercial-scale plant is a difficult task [6].

To protect the environment and reduce greenhouse gas emissions, methods of obtaining biogas and biohydrogen using alcohol waste and spent coffee grounds were developed. The disposal of these wastes can increase the efficiency and profitability of biofuel production in the future. Untreated waste contributed to the increased production of methane and biogas, while pre-treated waste produced more biohydrogen [7,8].

In the second direction, the problem of finding ways to use the products of alcohol production emerges acutely. It is known that in the process of alcohol production, in addition to ethanol, a number of by-products are obtained, among which are distillers' residue, final stage stillage, the main fraction, fuel oil, and fuel alcohol. The non-volatile part of the brew is removed with the distillers' residue and final stage stillage; volatile impurities are removed with the main fraction or its concentrate, with fuel oil and fuel alcohol.

The expansion of the alcohol industry in the world necessitated the development of effective technologies for the processing of the alcohol distillers' residues. The amount of the distillers' residue, which is formed during the processing of grain, is about 12 times higher than the amount of produced alcohol. Due to the low content of dry matter (up to 8%), the short shelf life (up to 2 days), the seasonality of consumption in animal husbandry, and the peculiarities of the distillers' residue transportation, there is a need for its disposal. Due to the significant proportion of protein in the raw material from which distillers' residue comes, it contains a large amount of total nitrogen and phosphorus. Therefore, due to the significant concentration of organic substances and the low pH value, the discharge of a large number of liquid effluents containing alcohol distillers' residue can cause soil pollution. The authors propose a combined technology for the treatment of such effluents with the valorization of alcohol distillers' residue, which is a valuable source of polysaccharides and volatile fatty acids, as well as natural antioxidants, including polyphenols and other bioactive compounds. All this may be of interest to the pharmaceutical, cosmetic, and food industries. It has been documented that for effective degradation of organic matter, particularly from the point of view of water reuse, complex treatment involving several sequential technologies should be applied. Special attention is paid to the consideration of new eutectic solvents for extracting the mentioned compounds [9]. There are well-known methods of cleaning such wastewater using membranes and materials that reduce membrane pollution [9,10].

It is interesting to study the use of distillers' residue to obtain pectins. Isolated pectins from distillers' residue show astringent properties, due to which the content of acidic components of gastric juice is normalized, which is important for their use in medical nutrition [11].

It is known that the starch distillers' residue shows a tendency to acid fermentation. This is due to the formation of organic acids for a relatively short period, mainly lactic acid, which usually dominate in this type of distillers' residue [12].

One of the waste products of the sugar-alcohol agribusiness process is filter sediment, which contains organic substances, mainly proteins and lipids, rich in calcium, nitrogen, potassium, and phosphorus. It has been shown that such filter sediment can be an effective adsorbent in polluted environments for the removal of potentially toxic metals (Cu(II), Cd(II), Pb(II), Ni(II), and Cr(III)) [13].

Among the by-products of ethanol production, the use of fuel oil can be of interest. Fuel oil is an intermediate product that is released at the stage of rectification from the clove column, the composition of its components depending on the type of raw material, as well as on the peculiarities of the technological process. Fuel oil contains a mixture of alcohols, water, and a small amount of other organic compounds [14]. There is a well-known study on the use of fuel oil for the production of ethers [14].

Our goal was to investigate the possibility of using clove oil and clove alcohols in the fuel industry to improve the operational properties of motor fuels, as well as to determine the technological parameters of creating effective fuel mixtures.

Experimental

Commercial gasoline of the A-78 brand, a straight-run fraction of diesel fuel after atmospheric distillation, and fuel oil (FO) (a waste product of alcohol production) were taken for experimental research. Fuel oil contains a mixture of higher alcohols (isoamyl, isobutyl, n-propyl, and ethyl), water, and a small number of esters. Fuel alcohols (FA) were obtained from fuel oil by distillation from water.

The research was carried out in two following stages: the study of the influence of FO and FA on the quality of gasoline (1st stage) and the study of the influence of FO on the quality of diesel fuel (2nd stage). At the first stage, mixtures of gasoline with additives of fuel oil (FO) and fuel alcohol (FA) were prepared in the following volume proportions:

1. Gasoline (95%)+FA (5%),
2. Gasoline (95%)+FO (5%),
3. Gasoline (92.5%)+FA (7.5%),
4. Gasoline (92.5%)+FO (7.5%),
5. Gasoline (90%)+FA (10%),
6. Gasoline (90%)+FO (10%),
7. Gasoline (85%)+FA (15%),
8. Gasoline (85%)+FO (15%).

Distillation of gasoline and prepared mixtures was carried out, during which their fractional composition was determined, namely: the temperature of the beginning of boiling, the boiling points of the 10%, 50%, and 90% points, and the temperature of the end of boiling [15]. The resulting fractional compositions were analyzed and octane numbers were determined for pure gasoline and prepared mixtures.

Octane number (ON) is a conditional quantitative characteristic of the fuel's ability to detonate, which characterizes its detonation resistance, and was determined by an analytical and computational method. This method involves the calculation of the ON of gasoline based on known values of relative density (ρ_4^{20}) and points of fractional composition (boiling temperatures of 10% ($T_{10\%}$) and 90% ($T_{90\%}$)) of gasoline) [15]. The octane number was calculated according to the following formula:

$$ON=1020.7-64.86[4\cdot lg(141.5/\rho_4^{20}-131.5)+2\cdot lg(1.8T_{10\%}+32)+1.3\cdot lg(1.8T_{90\%}+32)].$$

The densities of gasoline and mixtures were determined by the pycnometric method [14]. The molecular weight of gasoline and mixtures was determined by the average boiling point:

$$T_{av}=(T_{sb}+T_{10\%}+T_{50\%}+T_{90\%}+T_{eb})/5.$$

At the second stage, a mixture of diesel fuel (DF) was prepared with additives of fuel oil (FO) in the following volume proportions:

1. DF (95%)+FO (5%),
2. DF (92%)+FO (8%),
3. DF (90%)+FO (10%),
4. DF (85%)+FO (15%).

The density (ρ^{15}) of the resulting mixtures was determined by the pycnometric method, and the kinematic viscosity (ν) was determined by a capillary viscometer. After that, diesel fuel and prepared mixtures were distilled, during which their fractional composition was determined: the temperature at the beginning of boiling, and the boiling point of the 10% and 50% points. The resulting fractional compositions were analyzed and cetane numbers of the analyzed samples were determined.

Cetane number (cetane index, CI) is an indicator of diesel fuels characterizing the ignition delay period from the compression of the fuel-air mixture. This indicator is used in European standards. Determination of CI was carried out according ISO 4264:1995, IDT by the method that consists in determining the density of diesel fuel at 15^0C and the average boiling temperature of 50 vol.% of its quantity. The cetane index was calculated according to the formula:

$$CI=454.74-1641.416\cdot\rho+774.74\cdot\rho^2-0.554\cdot T_{50\%}+97.803(lgt)^2,$$

where ρ is the density of diesel fuel at 15°C (g/cm³); and $T_{50\%}$ is the boiling temperature of 50 vol.% of fuel (°C).

Results and discussion

At the first stage of research, the influence of fuel oil and fuel alcohol on the density, fractional composition, and detonation resistance of gasoline was established to analyze how the nature of fuel combustion and operational properties of gasoline will change.

The experimental results showed (Table 1) that the addition of fuel alcohol at lower concentrations more effectively affects the octane number of gasoline; the optimal concentration of FO is 5 vol.%, at which the ON of the fuel mixture increases by 10 units. Activating ability of fuel oil to increase the octane number manifests itself after adding them to gasoline in an amount of more than 6 vol.%; the optimal concentration is 10 vol.%, at which the ON of the fuel mixture increases by 1.5–2 units. As the proportion of FO and FA in the fuel mixture increases above 10 vol.%, the octane number begins to decrease.

To evaluate the operational properties of the prepared fuel mixtures, their fractional composition was analyzed. Based on the results of the analysis (Figs. 1 and 2), the following conclusions can be drawn.

With the addition of alcohol products, the temperature of the beginning of boiling increases, which reduces the tendency of the fuel to form vapor-air plugs in the engine power system. Fuel alcohols more effectively affect this indicator (Fig. 1), especially, at the concentration of 5 vol.%, fuel oil increases the boiling point (Tsb) to a lesser extent, but shows a more stable and permanent effect (Fig. 2).

With the addition of fuel alcohol, the boiling point of 10% of the fuel is lowered (Fig. 1), which improves the starting properties of the engine in cold conditions and its tendency to the formation of gas plugs in the system, while the addition of fuel oil practically does not affect this indicator (Fig. 2).

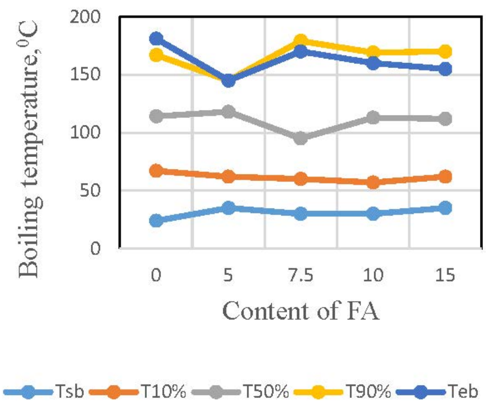

Fig. 1. Dependence of the boiling point of the fractional composition of gasoline on the amount of the addition of FA fuel alcohols

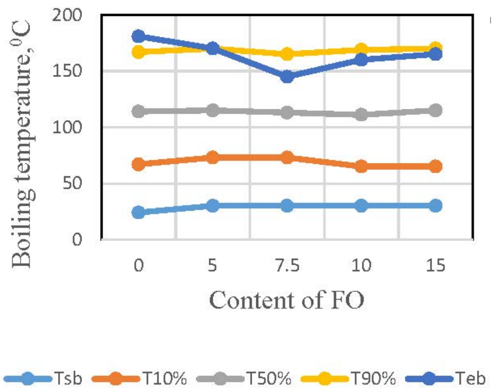

Fig. 2. Dependence of the boiling point of the fractional composition of gasoline on the amount of supplement of FO fuel oil

With the addition of fuel oil and especially fuel alcohol, the boiling point of 50% of gasoline is lowered, which facilitates its evaporation and the rapid transition of the engine from one operating mode to another, as well as the uniform distribution of fuel among the cylinders. The optimal concentrations of

Table 1

The main operational parameters of a gasoline-based fuel mixture with additives of fuel oil (FO) and fuel alcohol (FA)

The content of the fuel mixture, vol.%			Main characteristics of the fuel mixture							
Gasoline	FA	FO	ρ_4^{20}	T_{sb}, °C	$T_{10\%}$, °C	$T_{50\%}$, °C	$T_{90\%}$, °C	T_{eb}, °C	M	ON
100	–	–	0.7680	24	67	114	167	181	217	78.3
95	5	–	0.7711	35	62	118	145	145	192	88.6
95	–	5	0.7718	30	73	115	170	170	207	78.04
92.5	7.5	–	0.7696	30	60	95	179	170	207	82.02
92.5	–	7.5	0.7707	30	73	113	165	145	219	78.70
90	10	–	0.7681	30	57	113	169	160	204	85.40
90	–	10	0.7595	30	65	111	169	160	207	79.60
85	15	–	0.7644	35	62	112	172	155	209	79.60
85	–	15	0.7718	30	73	115	170	175	207	78.75

additives are as follows: 5–8 vol.% (Fig. 1) and 10 vol.% (Fig. 2) for FA and FO, respectively.

With the addition of fuel alcohol and fuel oil, the boiling point of 90% and the end of boiling of the fuel are lowered, which has a positive effect on the completeness of its combustion and the uniformity of its supply to the engine cylinders. The optimal concentrations of additives are as follows: 5–7.5 vol.% (Fig. 1) and 7.5–10 vol.% (Fig. 2) for FA and FO, respectively. When adding alcohol additives, the fuel density does not increase significantly, and therefore there are no problems with the homogeneity of the working mixture and its pumping in the engine system (Table 1).

Having analyzed the effect of both additives on the ON and the fractional composition of gasoline, one can conclude that the optimal concentrations of alcohol additives to gasoline are as follows: 5 vol.% and 10 vol.% vol for FA and CO, respectively. Figures 3 and 4 show comparison diagrams of the operational characteristics of fuel mixtures at the optimal concentrations of FA and FO.

At the second stage of research, the effect of fuel oil on the fractional composition and cetane index of diesel fuel was determined (Table 2).

The given data show that fuel oil additives increase the cetane index of diesel fuel in the interval observed for FO concentration from 5% to 10% (Fig. 5). In other words, the additive has a positive effect on the CI indicator, because the greater the value of the cetane index, the faster its preliminary oxidation will occur in the combustion chamber, and the faster the mixture will ignite and the engine will start. A particularly noticeable effect is observed in the concentration range of FO additives of 8–10 vol.%; there is a stabilization of the CI when the concentration of FO increases above 10 vol.% (Fig. 5).

An important characteristic of a diesel fuel is its viscosity. As can be seen in Fig. 6, the viscosity of the fuel mixture decreases with the addition of FO, which has a positive effect on the completeness of mixing and its transportation to the engine's fuel system.

To evaluate the operational properties of the prepared samples of diesel fuel mixtures, their fractional compositions were analyzed (Table 2, Fig. 7). Based on the results of the analysis, the following conclusions can be drawn. With the addition of FO, the temperature of the beginning of boiling, which characterizes the starting properties of the fuel, decreases. As this temperature decreases, the amount of substances that evaporate easily increases, and this makes it possible to start the engine more easily and at a lower temperature of the surrounding air. When FO additives are increased, the temperature at the boiling point of 10% of the fuel mixture decreases, which also improves the starting properties of the engine (Fig. 7).

As for the effect of FO additives on the boiling point of 50% of the amount of fuel, a rather significant decrease of this indicator is observed in the previously mentioned range of FO concentrations of 8–10 vol.% (Fig. 7). This has a positive effect on the diesel engine because fuel evaporation is facilitated, the possibility of smooth and stable operation of the engine is ensured, its maneuverability is significantly improved, and fuel consumption is also reduced.

The optimal concentration of FO additive to DP, at which the best fuel characteristics are observed, can be taken as 10 vol.%. Figure 8 shows a diagram comparing the fractional composition of fuel mixtures at the optimal FO concentration.

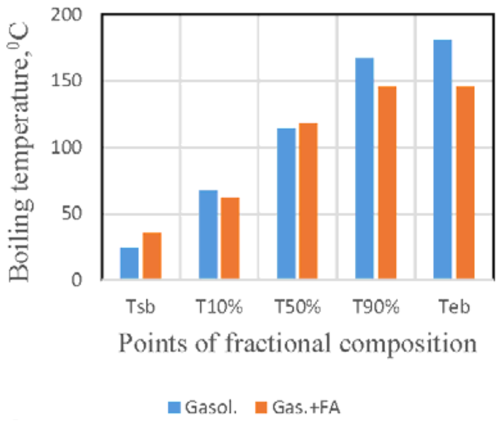

Fig. 3. Comparative characteristics of the boiling temperatures of the fractional composition of gasoline and the gasoline mixture with the addition of fuel alcohols FA at the content of 5 vol.%

Table 2

The main operational parameters of a fuel mixture based on diesel fuel with additives of fuel oil (FO)

The content of the fuel mixture, vol.%		Main characteristics of the fuel mixture					
Diesel fuel	FO	ν^{20}, sSт	ρ^{15}, kg/m^3	T_{sb}, ^{0}C	$T_{10\%}$, ^{0}C	$T_{50\%}$, ^{0}C	CI
100	–	4.4822	835.7	194	233	288	41.33
95	5	4.4048	834.3	152	198	285	42.44
92	8	4.3221	833.2	110	190	280	43.41
90	10	4.2829	833.4	116	186	287	45.72
85	15	4.2534	832.6	110	174	285	45.95

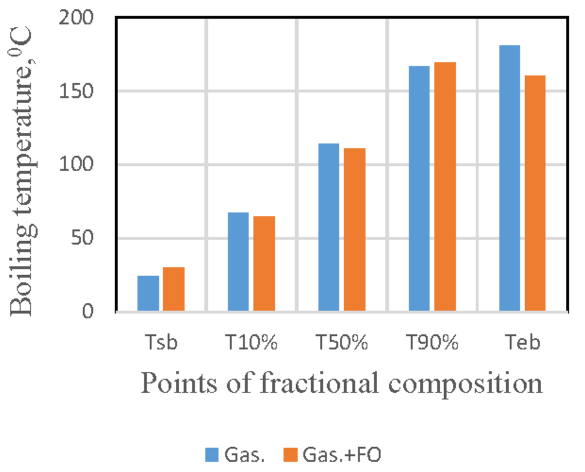

Fig. 4. Comparative characteristics of the boiling temperatures of the fractional composition of gasoline and gasoline mixture with the addition of fuel oil FO at the content of 10 vol.%

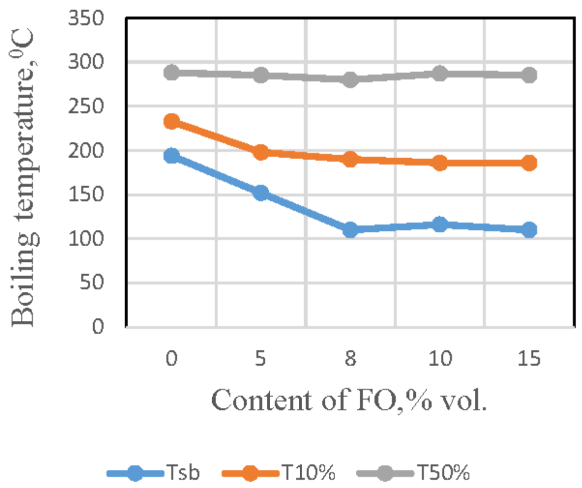

Fig. 7. Dependence of the boiling temperatures of the fractional composition of diesel fuel on the amount of fuel oil additive FO

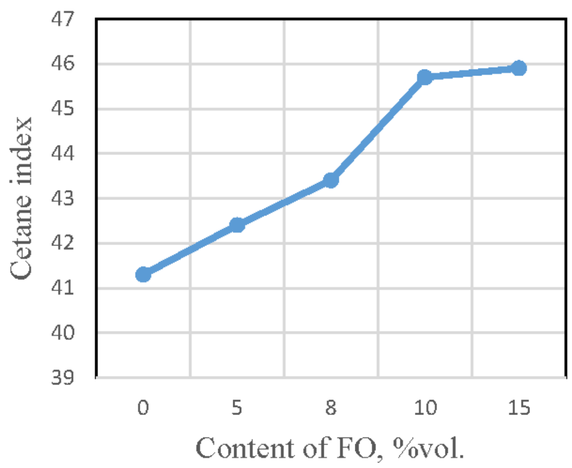

Fig. 5. Dependence of the cetane index of diesel fuel on the amount of fuel oil additive FO

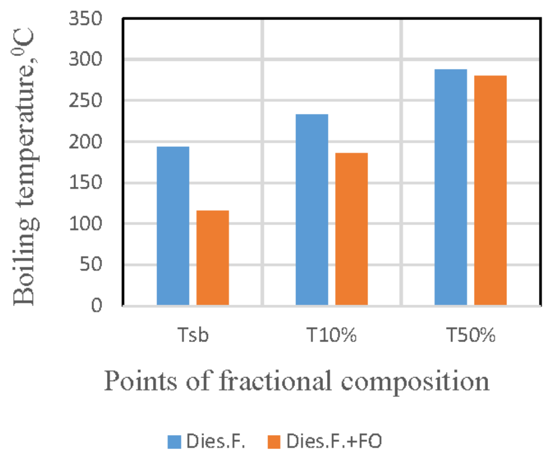

Fig. 8. Comparative characteristics of the boiling temperatures of the fractional composition of diesel fuel and the fuel mixture with the addition of fuel oil at the content of 10 vol.%

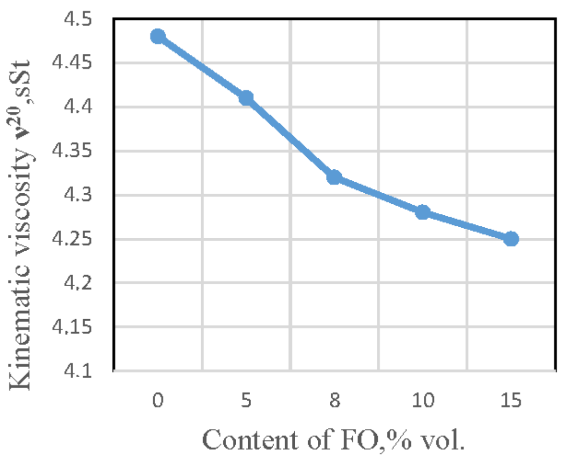

Fig. 6. Dependence of the viscosity of diesel fuel on the amount of fuel oil additive FO

Conclusions

The by-products of ethanol production, fuel alcohol and fuel oil, can be successfully used in the fuel industry.

Alcoholic products improve the combustion characteristics of gasoline, increase their detonation resistance, and improve their operational properties. When added to gasoline, fuel alcohols, which are freed from water and organic impurities, are more effective. The optimal concentrations of additives are as follows: 5 vol.% and 10 vol.% for fuel alcohol and fuel oil, respectively.

The addition of fuel oil to diesel fuels increases their cetane number, has a positive effect on the fractional composition and viscosity of the fuel mixture and, in general, improves the operational properties of the fuel. The optimal value of the concentration of the content of the alcohol additive is 10 vol.%.

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ЗАСТОСУВАННЯ ПОБІЧНИХ ПРОДУКТІВ ВИРОБНИЦТВА ЕТАНОЛУ У ПАЛИВНІЙ ПРОМИСЛОВОСТІ

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В даній статті проаналізовано два напрями вирішення екологічних проблем у паливній промисловості, які виникли для необхідності стабілізації концентрації парникових газів в атмосфері. За першим напрямом розглянуто способи використання різних нетрадиційних видів сировини і відходів, що містять цукри, з метою виробництва екологічно чистих видів палива. За другим напрямом аналізуються можливості комплексного використання етанолу та усіх побічних продуктів, які утворюються при його виробництві, для покращення екології в паливній промисловості. Запропонована можливість використання побічних продуктів виробництва етанолу (сивушної олії і сивушного спирту) для покращення експлуатаційних властивостей моторних палив, які застосовуються в карбюраторних і дизельних двигунах. Показано, що добавки сивушного спирту і сивушної олії підвищують октанове число бензину, покращують його фракційний склад, в результаті чого збільшується повнота згоряння палива та рівномірність її подачі в циліндри двигуна. Оптимальними концентраціями спиртових добавок до бензинів є такі: для сивушних спиртів – 5 об.%, для сивушної олії – 10 об.%. Встановлено, що додавання сивушної олії до дизельних палив підвищує їх цетановий індекс, у зв'язку з чим швидше загоряється суміш та запускається дизельний двигун. Крім цього з додаванням сивушної олії знижується температура початку кипіння, що полегшує пускові властивості палива, а також зменшується в'язкість паливної суміші, що покращує повноту змішування і транспортування в системі двигуна. Оптимальною концентрацією добавки сивушної олії до дизельного палива є значення 10 об.%.

Ключові слова: сивушна олія, сивушні спирти, октанове число, бензин, цетановий індекс, дизельне паливо.

APPLICATION OF BY-PRODUCTS OF ETHANOL PRODUCTION IN THE FUEL INDUSTRY

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This article analyzes two ways of solving environmental problems in the fuel industry, which arose due to the need to stabilize the concentration of greenhouse gases in the atmosphere. According to the first direction, the ways of using various non-traditional types of raw materials and waste containing sugars for the purpose of producing environmentally friendly types of fuel are considered. According to the second direction, the possibilities of integrated use of ethanol and all by-products formed during its production are analyzed to improve ecology in the fuel industry. We have established the possibility of using the by-products of ethanol production, fuel oil and fuel alcohol, to improve the operational properties of motor fuels used in carburetor and diesel engines. It has been stated that the additives of fuel alcohol and fuel oil increase the octane number of gasoline, improve its fractional composition, as a result of which the completeness of fuel combustion and the uniformity of its supply to the engine cylinders increase. The optimal concentrations of alcohol additives to gasoline are as follows: 5 vol.% and 10 vol.% for fuel alcohols and fuel oil, respectively. It has been established that the addition of fuel oil to diesel fuels increases their cetane number, this causes the mixture to ignite faster and the diesel engine to start. Moreover, with the addition of fuel oil, the temperature of the beginning of boiling decreases which facilitates the starting properties of the fuel, and the viscosity of the fuel mixture decreases which improves the completeness of mixing and transportation in the engine system. The optimal concentration of fuel oil additive to diesel fuel is 10 vol.%.

Keywords: fuel oil; fuel alcohol; octane number; gasoline; cetane number; diesel fuel.

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