

УЛУЧШЕНИЕ НИЗКОТЕМПЕРАТУРНЫХ СВОЙСТВ ДИЗЕЛЬНОГО ТОПЛИВА С ПОМОЩЬЮ ДЕПРЕССОРНЫХ ПРИСАДОК

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Статья посвящена обзорному анализу веществ, обладающих депрессорным действием по отношению к дизельному топливу. Для обеспечения производства дизельных топлив с улучшенными низкотемпературными свойствами в необходимом объеме могут применяться различные методы. Одним из наиболее перспективных методов является введение в топливо веществ, обладающих депрессорными свойствами в виде присадок. Их действие основано на снижении значения предельной температуры фильтруемости и температуры застывания после введения присадки в сравнении с исходным топливом. Приведен анализ различных групп и видов депрессоров. В зависимости от строения депрессорного компонента присадок различаются технологии их производства, а также степень влияния на дизельное топливо. Для отдельных видов присадок известны экспериментальные значения рабочих концентраций в топливе и степени депрессий различных показателей низкотемпературных характеристик дизельного топлива, а также приведены их структурные формулы. Из приведенного анализа видно, что и исследования полимерных соединений в качестве компонентов депрессорных присадок становятся более разнообразными, и результаты этих исследований широко внедряются в производство. Приведен анализ основных современных концепций механизма действия депрессорных присадок на дизельное топливо. Среди наиболее распространенных можно выделить две концепции механизма, а именно адсорбционный механизм и сокристаллизационный механизм. Также приведен анализ современного мирового рынка депрессорных присадок. Основными поставщиками депрессорных присадок являются зарубежные компании, а именно Clariant, BASF, Infenium International. Данные компании выпускают широкий ассортимент депрессорных присадок для дизельных топлив на основе различных сополимеров. Кроме того, среди отечественных поставщиков также имеются линейки депрессорных присадок, представленных широким спектром полимерных соединений.

Ключевые слова: дизельное топливо, депрессорные присадки, низкотемпературные свойства

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IMPROVING THE LOW-TEMPERATURE PROPERTIES OF DIESEL FUEL WITH THE HELP OF POUR-POINT DEPRESSANTS

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The article is devoted to a general review of substances that have a depressant effect in relation to diesel fuel. To ensure the production of diesel fuels with enhanced low-temperature properties in the required volume, various methods can be used. One of the most promising methods is the introduction of substances with depressant properties into the fuel in the form of additives. Their action is based on a decrease in the value of the limiting filterability limit temperature and pour point after the introduction of the additive in comparison with the original fuel. An analysis of various groups and types of depressants is provided. Depending on the structure of the depressant component of additives, there are various technologies for their production, as well as the degrees of effect on diesel fuel. For certain types of additives, there are experimental values of use rate in the fuel and the degree of depression of various indicators of low-temperature characteristics of diesel fuel. Their structural formulas are also given. The analysis shows that the production and research of polymer compounds as components of pour-point depressants is becoming more profound and diverse. An analysis of the main modern concepts of the mechanism of action of pour-point depressants on diesel fuel is given. There are two most common mechanism concepts, namely the adsorption mechanism and the co-crystallization mechanism. An analysis of the modern world market of pour-point depressants is also provided. The main suppliers of pour-point depressants are foreign companies, namely Clariant, BASF, Infenium International. Among domestic suppliers there are also lines of pour-point depressants, represented by a wide range of polymer compounds used as pour-point depressants.

Key words: diesel fuel, pour point depressant, low temperature properties

Diesel fuel is one of the most important types of fuels [1]. In 2021, 80395 thousand tons of diesel fuel were produced in the Russian Federation, which is 3.3% more compared to the previous year [2]. It is expected that by 2025 the share of diesel fuel in the structure of consumption of petroleum products will reach 37%. The largest consumer of diesel fuel is the transport sector [3], in particular, agricultural machinery. Based on the data [4], the consumption of diesel fuel in agriculture exceeds the consumption of gasoline more than 5.5 times.

There is a number of requirements imposed on diesel fuel, in particular, related to their low-temperature properties. The presence of a significant amount of alkanes in diesel fuel, especially of normal structure, is one of the reasons for the deterioration of such low-temperature characteristics as cloud point, filterability

limit temperature, and chilling point [5]. Taking into account the features of Russian climate, the problem of regulating the low-temperature properties of diesel fuel is of particular importance. It is also worth noting that the existing capacities of refineries for the production of winter and arctic diesel fuels are not enough given the continuously growing demand for these types of fuels [6].

In Russia, there are several standards that regulate the properties of diesel fuel. All-Union State Standard (GOST) 52368-2005 [7] is similar to the European standard EN 590 for diesel fuels and provides for production of diesel fuels in Russia in accordance with European standards. The specified standard provides for the production of six grades of diesel fuel (A, B, C, D, E, F), which differ from each other in low-temperature properties, characterized by the filterability limit temperature index. Also in [7], fuel for cold

and arctic climates is divided into classes 0, 1, 2, 3, and 4, which differ from each other in terms of the filterability limit temperature and cloud point. According to the All-Union State Standard (GOST) 32511-2013 [8], diesel fuels are divided into summer, off-season, winter, and arctic types.

In case diesel fuel does not fit in with the established standards in terms of the low-temperature characteristics, it is necessary to introduce processes that improve cloud point and filterability limit temperature in the technology of its production. The following methods are used for this:

- 1) decrease in the end of the boiling of the diesel fraction;
- 2) mixing diesel fraction with kerosene fraction;
- 3) removal of n-paraffins by extractive crystallization;
- 4) catalytic isomerization;
- 5) catalytic dearafinization;
- 6) introduction of pour-point depressants [9].

By adding pour-point depressants to the fuel it is possible to reduce the filterability limit temperature and the temperature of the pour point [1, 10].

Pour-point depressants are chemicals that, due to their physicochemical properties, can affect the process of crystallization of paraffins. The use rate of additives is 0.02-0.10 %, which makes this improvement method more economical [9, 11].

Pour-point depressants can be classified in the following way [1]:

- polymer compounds;
- organic non-polymer compounds.
- In turn, polymeric compounds include the following substances [1, 13]:
 - copolymers of ethylene with polar monomers;
 - polymers based on esters of acrylic and methacrylic acids;
 - polyolefins;
 - copolymers of maleic anhydride with alpha-olefins;
 - alkylphenol-formaldehyde resins.

Non-polymeric organic compounds acting as depressants are as follows [1, 13, 14, 15]:

- alkylnaphthalenes;
- esters of polyhydric alcohols;
- esters of dibasic acids;
- nitrogen-containing compounds
- nanohybrid additives
- oil refinery residues.

Most of the additives produced are polymer compounds. Among this class of additives, copolymers of ethylene with polar monomers are widespread.

Compounds based on the copolymer of ethylene with vinyl acetate are a well-known depressant. Monomeric links in the polymer chain are statically distributed. A wide variety of polymer properties can be achieved by varying the ratio of monomers and the molecular mass of the molecule, which can achieve 103-105 g/mol. The content of vinyl acetate links in the molecule is preferably 20-40% [16]. The number of methyl branches from the main chain can be estimated on average as 8.5 per 100 methylene groups. The disadvantage of ethylene-based compounds is the strict conditions of their synthesis: up to 100 MPa, 100-150 °C [1, 17].

The scheme of copolymer ethylene and vinyl acetate production is shown in Fig. 1.

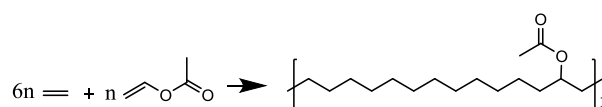


Fig. 1. Scheme of copolymer ethylene and vinyl acetate production
Рис. 1. Схема получения сополимера этилена и винилацетата

According to experimental data, it is known that at working concentrations of 0.01-0.10% by volume, the depression of the pour point and filterability limit temperature are up to 30 and up to 15 °C, respectively. The cloud point remains the same in this case [13, 18].

It was also experimentally found that ethylene-styrene copolymers have a depressant effect on diesel fuel. In this case, it is possible to achieve the production of winter and off-season fuel from the summer basis [19].

A large group of compounds are copolymers based on esters of acrylic and methacrylic acids. A common feature of additives of this type is the structure of the alkyl(meth)acrylate monomer. The molecule of such an ester has a linear hydrocarbon radical with an average length of 12 to 22 carbon atoms. It is in this interval that the optimal depressant activity of the additive is manifested [1].

There are additives based on copolymers of alkyl(meth)acrylate and ethylene or alpha-olefins. Additives of this type are capable of lowering the chilling point of fuel to -39 °C, and filterability limit temperature to -20 °C [1].

The structure of the copolymer of (meth)acrylic acid esters and alpha-olefin is shown in Fig. 2.

The properties of copolymers of alkyl methacrylates and styrene have been well studied. 0.1-0.5% of this additive in diesel fuel makes it possible to achieve an improvement in the low-temperature characteristics of the fuel [20].

Copolymers of alkyl methacrylates and maleic anhydride have little effect on the properties of diesel fuel. However, after the modification of the maleic anhydride monomer by obtaining amides and imides, the low-temperature properties of diesel fuel are significantly improved [21].

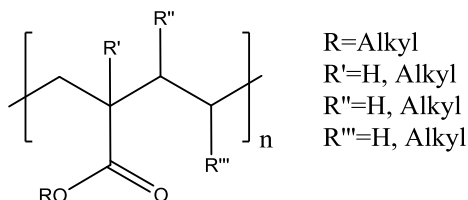


Fig. 2. Structure of copolymer of (meth)acrylic acid esters and alpha-olefin

Рис. 2. Структура сополимера эфиров (мет)акриловой кислоты и альфа-олефина

Unlike additives based on ethylene, in the production of alkyl(meth)acrylate additives of this type, quite mild conditions are used, such as atmospheric pressure and temperatures below 100 °C [22].

The simplest structure among polyolefins with depressant properties is low molecular weight branched polyethylene, which, with an average molecular weight of 1800-4200 g/mol, makes it possible to achieve a decrease in the chilling point to -38 °C, and filterability limit temperature to -23 °C [1].

Branched polyolefins based on ethylene and propylene monomers also improve the low-temperature properties of diesel fuel. To obtain such compounds, harsh conditions are necessary: the pressure up to 300 MPa and the temperature of 200 °C. Otherwise, a depressant compound can be obtained by destruction of ethylene-propylene rubber [13].

The structure of the block copolymer of ethylene and propylene is shown in Fig. 3.

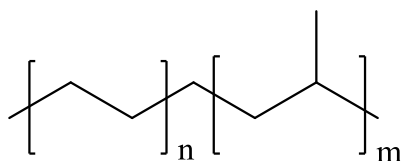


Fig. 3. Structure of the block copolymer of ethylene and propylene
Рис. 3. Структура блок-сополимера этилена и пропилена

Modified copolymers of maleic anhydride and alpha-olefins can be used as a depressant component of additives. The use rate of the copolymer is 0.05-0.10%. In this case, it is possible to achieve a depression of filterability limit temperature by 20-25 °C. Maleic anhydride units are modified by converting them into the form of esters, imides or amides (see Fig. 4).

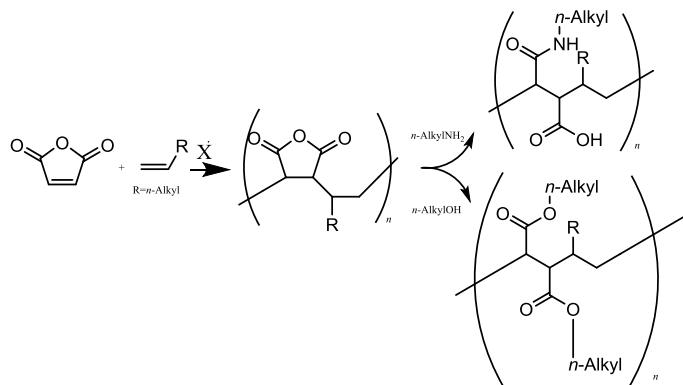


Fig. 4. Reactions for obtaining derivatives of copolymers of maleic anhydride with alpha-olefins

Рис. 4. Реакции получения производных сополимеров малеинового ангидрида с альфа-олефинами

Alkylphenol-formaldehyde resins are produced in two stages. The first step is the phenol alkylation reaction, the Friedel-Crafts reaction. At the second stage, the resulting alkylphenol reacts with para-formaldehyde, resulting in a polymer resin with depressant properties [13].

The structure of the alkylphenol-formaldehyde resin is shown in Fig. 5.

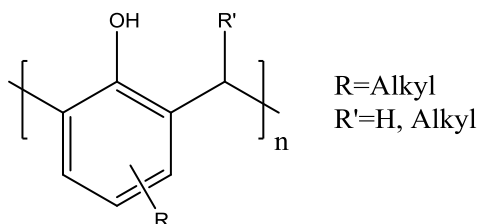


Fig. 5. Structure of the alkylphenol-formaldehyde resin
Рис. 5. Структура алкилфенолформальдегидной смолы

Unsaturated etheronaphthalenes with a molecular weight of about 1200 g/mol can be used as components of depressant compounds. This substance is obtained by condensation of naphthalene and an ester of oleic acid and spermaceti alcohols [1].

The structure of etheronaphthalene is shown in Fig. 6.

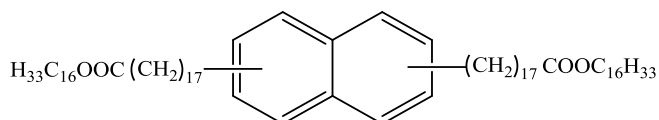


Fig. 6. Structure of etheronaphthalene
Рис. 6. Структура эфиринафталина

The non-polymeric compounds may also be esters of polyhydric alcohols such as glycerol, mono-, di-, triethylene glycol. In addition, pentaerythritol oxytearate is capable of producing a depressant effect [12].

Nitrogen-containing non-polymer components of additives are most often dialkylamines and alkylamides. One of the substances of this type is dialkylcyanamide obtained by the condensation of potassium cyanamide with haloalkyls [1].

In addition, some oil refinery residues, in particular, the oxidation product of heavy pyrolysis tar, can be used as a depressant-active component [13].

Depressant molecules interact with paraffin crystals. Currently, there is no consensus on the mechanism of action of depressants. However, there are two points of view that are the most common [10].

The adsorption mechanism involves the sorption of the active component of the additive on the surface of the paraffin crystal with the help of the polar part. In this case, the nonpolar part of the depressant is facing the hydrocarbon medium and prevents the approach of crystals and their association into a space framework [24].

According to the cocrystallization mechanism, the nonpolar part of the active substance is incorporated into the paraffin crystal, and the polar part of the depressant molecule prevents the deposition of new paraffin molecules on the existing crystal, which could lead to its further growth. It is also assumed that the depressant affects the volume and surface of n-paraffin crystals [10].

The effectiveness of the additive depends on the hydrocarbon contained in diesel fuel. Namely, depression is affected by the presence of such groups of compounds as n-paraffins, monocyclic and polycyclic aromatic hydrocarbons. For this reason, the same pour-point depressant can have different effects on the fuel.

An increase in the presence of paraffins in the fuel adversely affects both the low-temperature properties of the fuel and the efficiency of the additive. At the same time, an increase in the relative proportion of high-boiling paraffins has a positive influence on the effect of the additive. This is due to the rate of crystal formation. In fuels with a narrow distribution of paraffins, crystals form rapidly, and the depressant molecules do not have time to interact with the crystals. In fuels with a wide composition of paraffins, crystal formation is carried out more slowly and in a wider temperature range, which allows the additive to interact with the crystal growth process. Thus, the effect of pour-point depressants on diesel fuel is higher with a wider fractional composition [25, 26].

Studies have shown that a high content of monocyclic aromatic hydrocarbons has a negative impact on the depressant effect of the additive. This is due to the high intake capacity of aromatics to the polymer additive. Due to their polarity, aromatic hydrocarbons

adsorb depressant molecules on themselves. In this case, the effectiveness of the additive with respect to paraffin crystals decreases. For aromatic compounds, as the number of cycles increases, the intake capacity to the additive decreases, and the effect of the polymer on crystal growth does not decrease. Therefore, the greater the average number of aromatic cycles in the fuel, the higher the efficacy of the additive [25, 27].

The modern diesel fuel industry that meets quality standards requires a wide range of pour-point depressants on the market. This is due to the fact that depressants are not universal additives for diesel fuel. At each refinery that produces diesel fuel, the type of additive is often experimentally selected [24]. It is important to note that the European pour-point depressant market is shaped mainly by such companies as Clariant, BASF and Infineum [24, 28].

BASF produces more than 30 brands of additives, among which 5 are depressant ones: Keroflux ES 3566, 3579, 3580 [29], 3501, 3520 [28]. The previously mentioned additives are obtained by copolymerization of ethylene and vinyl esters or ethylene and acrylic acid esters [24]. Keroflux 3501, 3520, 3566 additives are used at four Russian oil refineries [28].

Clariant offers the following brands of pour-point depressants: Dodiflow 4741, 4603, 4971 and their analogues [24]. Commercial forms of additives of these lines are a mixture of ethylene polymer with vinyl acetate and modified polymers in high-boiling hydrocarbon solvents. Additive lines Dodiflow 4851, 4965, 4971, 5057, 5416, 5747 are used at thirteen Russian refineries as a pour-point depressant [28].

The additive named Infineum offered by Infineum company (brand Infineum R 430) is used by one oil refinery [28-30].

The following domestic additives are known: VES-238, Polypren, VES-410D, PDP, Sandal-1B, EDEP-T, DAKS-D. VES-238, Polypren, VES410D, Sandal-1B are ethylenevinylacetate-based additives, while PDP and EDEP-T additives are based on copolymers of polyacrylates with vinyl acetate [10, 24]. DAKS-D is a pour-point depressant in which a low molecular weight copolymer of ethylene with alpha-olefins acts as an active component [10, 31].

CONCLUSION

One of the most important indicators of the quality of diesel fuels is its low-temperature characteristics. To ensure their compliance with European standards, there are various methods of improving the fuel, one of which is the introduction of pour-point depressants. This article discusses substances that improve the low-temperature characteristics of diesel

fuel. An analysis of the production of these compounds and the degree of their influence on the fuel is also provided. The main concepts of the mechanism of action of the additives are considered. Moreover, modern market of additives is analyzed.

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The authors declare the absence a conflict of interest warranting disclosure in this article.

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