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# Identification the Regularities of Influence of the Diesel Fuels Composition on the Efficiency of Low-Temperature Additives Action

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**Abstract**

The paper presents the results of determining the characteristics and composition of straight-run diesel fuels. In addition, low-temperature properties (cloud point, cold filter plugging point, pour point) the diesel fuels and low-temperature additives blends are studied. The regularities of influence the n-paraffins and aromatic hydrocarbons content in the diesel fuel composition on the effectiveness of the low-temperature additive is identified.

***Keywords****: Diesel fuel; Low-temperature properties; Additives; Group composition; Paraffins.*

## 1. Introduction

The harsh climate of the Russian Federation northern regions impose stricter requirements for diesel fuel (DF) operational characteristics. Winter and arctic brands of DF make it possible to effectively operate the equipment in the temperature range from -30°C to -50°C. Nowadays, the most effective way to obtain winter and arctic brands of DF that meet the requirements of [1] is the use of depressant additives.

The range of low-temperature additives – depressants and dispersants available on the market is diverse. As depressants, the widest copolymers of ethylene and vinyl acetate, copolymers of alkyl(meth)acrylates, copolymers of polyolefin type, copolymers of maleic anhydride etc. [2-9]. Also, nanohybrid additives are gaining popularity [10-11].

DF, in turn, also significantly differs in fractional, group, and hydrocarbon composition, depending on the oil field and the technology of petroleum products production [12].

The DF composition greatly affects the effectiveness of low-temperature additives, making it impossible to create universal compositions of additive and making it difficult to choose the optimal concentration of a particular additive. Thus, the aim of the work is to identify the regularities of influence the DF composition on the effectiveness of low-temperature additives.

## 2. Materials and methods

### 2.1 Object of study

The object of study in this paper is 3 samples of straight-run DF (assigned numerical codes from 1 to 3), as well as their blends with 6 low-temperature additives (assigned alphabetic codes). Additives were used in concentrations by manufacturer's recommended (Table 1).

Table 1. The concentration of used additives

| Additive concentration (by 100 mL sample), mL | | | | | |
|---|---|---|---|---|---|
| *A* | *B* | *C* | *D* | *F* | *G* |
| 0.100 | 0.231 | 0.300 | 0.620 | 0.100 | 0.260 |

### 2.2. Methods for determination of the DF samples characteristics

The studied samples of straight-run diesel fuel were characterized as follows: the density at the temperature of 15°C and 20°C was determined using the Stanbinger SVM3000 Anton

Paar viscometer according to the ISO 12185:1996 "Crude petroleum and petroleum products – Determination of density – Oscillating U-tube method" [13]. The viscosity at 20°C was determined using the Stanbinger SVM3000 Viscometer Anton Paar, according to the ISO 3104: 1994 "Petroleum products. Transparent and opaque liquids. Determination of kinematic viscosity and calculation of dynamic viscosity" [14]. The cetane index (*CI*) was determined according to the ISO 4264:2018 "Petroleum products – Calculation of cetane index of middle-distillate fuels by the four variable equation" [15-16]. The cloud point ($T_{cp}$) was determined according to the ASTM D2500-05 "Standard Test Method for Cloud Point of Petroleum Products" [17]. The cold filter plugging point (CFPP) was determined according to the ASTM D6371-17a "Standard Test Method for Cold Filter Plugging Point of Diesel and Heating Fuels" [18]. The pour point ($T_{pp}$) was determined according to the ASTM D97-17b "Standard Test Method for Pour Point of Petroleum Products" [19].

### 2.3. Methods for determining the composition of the DF samples

Fractional composition of the straight-run diesel fuel samples was determined according to the ISO 3405: 011 "Petroleum products – Determination of distillation characteristics at atmospheric pressure" [20].

The sulfur content was determined using the X-ray fluorescence energy dispersive analyzer "SPECTROSCAN S", according to the ASTM D4294-16 "Standard Test Method for Sulfur in Petroleum and Petroleum Products by Energy Dispersive X-ray Fluorescence Spectrometry" [21].

Structure-group composition of the diesel fuel samples was calculated using the n-d-M method according to the ASTM D3238-17a "Standard Test Method for Calculation of Carbon Distribution and Structural Group Analysis of Petroleum Oils by the n-d-M Method" [22].

To determine the group composition of the straight-run diesel fuel samples, the aniline method was used. The content of n-paraffins in the straight-run diesel fuel samples was determined by the gas-liquid chromatography method using the Chromatec-Crystal 2000 chromatograph with a quartz capillary column 30 m × 0.25 mm, stationary phase – SE-54, carrier gas – helium.

## 3. Results and discussion

### 3.1. Results of determining the DF samples characteristics

In the Russian Federation, four brands of DF are produced: S – summer, I – inter-season, W – winter, A – arctic [1]. Requirements of USS 305-2013 "Diesel fuel. Specifications" [1] are presented in Table 2.

Table 2. Requirements of USS 305-2013 "Diesel fuel. Specifications"

| Characteristic | Brand | | | |
|---|---|---|---|---|
| | *S* | *I* | *W* | *A* |
| Density at 15°C, g/cm$^3$, max | 863.4 | | 843.4 | 833.5 |
| Viscosity at 20°C, mm$^2$/s, max | 3.0 – 6.0 | | 1.5 – 5.0 | 1.5 – 4.0 |
| *CI* point, min | | | 45 | |
| $FC_{50\%}$,°C, max | | 280 | | 255 |
| $FC_{90\%}$, C, max | | | 360 | |
| Sulfur content, mg/kg, max | | | 2000 | |
| CFPP,°C, max | -5 | -15 | -25--35 | -45 |

The results of characteristics of straight-run DF samples determination are presented in Tables 3-4.

Table 3. The results of density, viscosity and *CI* determination

| DF sample | Density at 20°C, g/cm$^3$ | Density at 15°C, g/cm$^3$ | Viscosity at 20°C, mm$^2$/s | CI | $T_{CP}$, °C | CFPP, °C | $T_{PP}$, °C |
|---|---|---|---|---|---|---|---|
| 1 | 0.838 | 0.841 | 4.58 | 48.60 | -3 | -4 | -18 |
| 2 | 0.827 | 0.831 | 3.31 | 51.99 | -12 | -24 | -45 |
| 3 | 0.829 | 0.833 | 6.65 | 53.67 | -13 | -17 | -29 |

Table 4. The results of $T_{CP}$, CFPP and $T_{PP}$ determination

| DF sample | TCP, °C | CFPP, °C | TPP, °C |
|---|---|---|---|
| 1 | -3 | -4 | -18 |
| 2 | -12 | -24 | -45 |
| 3 | -13 | -17 | -29 |

According to the requirements of [1], the density of samples No. 2, 3 meet the requirements to all DF brands; sample No. 1 meets the requirements to S, I and W brands.

According to requirements of [1] by viscosity, sample No. 2 meets the requirements of all DF brands; sample No. 1 meets the requirements of brands S, I, W. Sample No. 3 does not meet the requirements of [1] by viscosity. All samples meet the requirements of [1] by CI.

According to the requirements of [1], by CFPP the samples No. 2, 3 meet the requirements of the brands W and I. Sample No. 1 does not meet the requirements of [1] by CFPP. Thus as the results show, it is impossible to obtain winter and, especially, arctic fuels without using the low-temperature additives.

### 3.2. The results of the determination of the DF samples composition

The results of the straight-run DF samples composition determination are shown in Tables 5-10.

Table 5. The results of fraction composition determination

| DF sample | *T*, °C | | | |
|---|---|---|---|---|
| | *IBP* | *V, mL* | | |
| | | *10* | *50* | *90* |
| 1 | 151 | 183 | 271 | 359 |
| 2 | 138 | 161 | 247 | 330 |
| 3 | 145 | 159 | 251 | 312 |

Table 6. The result of sulfur content determination

| DF sample | Sulfur content, mg/kg |
|---|---|
| 1 | 2517 |
| 2 | 1700 |
| 3 | 1710 |

According to the requirements of [1] by the fractional composition, samples No. 2, 3 meet the requirements of all DF brands; sample No. 1 meets the requirements of the brands S, I, W.

Samples No. 2, 3 meet the requirements of [1] by sulfur content. Sample No. 1 does not meet the requirements of [1] by sulfur content, and it needs desulphurization.

Table 7. Group composition of DF samples

| DF sample | Hydrocarbon content, % wt. | | |
|---|---|---|---|
| | *Aromatics* | *Naphthenic* | *Paraffins* |
| 1 | 24.14 | 31.24 | 44.63 |
| 2 | 23.82 | 29.02 | 47.16 |
| 3 | 22.82 | 32.57 | 44.61 |

The lowest content of aromatic hydrocarbons is typical for sample No. 3; the highest content of paraffin hydrocarbons is characteristic of sample No. 2, the lowest for sample No. 3.

Table 8. The results of n-paraffins content determination

| DF sample | N-paraffins content, % wt. | | |
|---|---|---|---|
| | *Total* | $C_{16+}$ | $C_{20+}$ |
| 1 | 19.61 | 8.88 | 4.01 |
| 2 | 22.05 | 8.34 | 3.04 |
| 3 | 19.88 | 7.14 | 2.32 |

The heaviest DF sample is sample No. 1 since this sample contains the largest number of n-paraffins $C_{20+}$, which explains it is the worst low-temperature properties; the sample No. 2 has the highest total content of n-paraffins; the sample No. 1 has the highest content of n-paraffins $C_{16+}$.

Table 9. Content of hydrocarbons ring in "average molecule"

| DF sample | Average number of rings in molecule | | |
|---|---|---|---|
| | $K_{ar}$ | $K_n$ | $K_t$ |
| 1 | 0.317 | 0.930 | 1.247 |
| 2 | 0.330 | 0.839 | 1.169 |
| 3 | 0.378 | 0.798 | 1.176 |

$K_{ar}$ – average number of aromatic rings; $K_n$ – average number of naphthenic rings;
$K_t$ – total average number of rings.

Sample No. 1 contains the largest average number of naphthenic rings in the molecule; sample No. 3 contains the largest average number of aromatic rings in the molecule. Generally, sample No. 1 characterized more ring structure.

Table 10. The results of DF samples n-d-M analysis

| DF sample | Carbon content, % wt. | | | |
|---|---|---|---|---|
| | $C_{ar}$ | $C_n$ | $C_{ring}$ | $C_{alk.chain}$ |
| 3 | 12.453 | 35.684 | 48.137 | 51.863 |
| 4 | 14.385 | 35.259 | 49.644 | 50.356 |
| 5 | 16.021 | 32.639 | 48.660 | 51.340 |

*$C_{ar}$ – hydrocarbon content in aromatic rings, % wt.; $C_n$ – hydrocarbon content in naphthenic rings, % wt.;*
*$C_{ring}$ – hydrocarbon content in rings structures, % wt.; $C_{alk.chain}$ – hydrocarbon content in alkyl chains, % wt.*

Sample No. 3 contains the largest proportion of carbon in aromatic rings; sample No. 1 – in naphthenic rings and alkyl chains.

### 3.3. Results of determination the low-temperature properties of DF and additives blends

Results of the low-temperature properties of DF sample and additives blends determination are presented in Tables 11-13.

Table 11. The change in $T_{CP}$ of DF samples with adding low-temperature additives

| DF sample | $T_{CP}$,°C | | | | | | |
|---|---|---|---|---|---|---|---|
| | *DF* | *Blend DF sample + additive* | | | | | |
| | | *A* | *B* | *C* | *D* | *F* | *G* |
| 1 | -3 | -5 | -6 | -4 | -3 | -2 | -4 |
| 2 | -12 | -12 | -18 | -12 | -13 | -12 | -13 |
| 3 | -13 | -15 | -16 | -15 | -16 | -13 | -16 |
| DF sample | $\Delta T_{CP}$ | | | | | | |
| | *av.* | *A* | *B* | *C* | *D* | *F* | *G* |
| 1 | 1.3 | 2 | 3 | 1 | 0 | 1 | 1 |
| 2 | 1.5 | 0 | 6 | 1 | 1 | 0 | 1 |
| 3 | 2.2 | 2 | 3 | 2 | 3 | 0 | 3 |
| $\Delta T_{CP}$ av. | | 1.3 | 4.0 | 1.3 | 1.3 | 0.3 | 1.7 |

Table 12. The change in CFPP of DF samples with adding low-temperature additives

| DF sample | CFPP, °C | | | | | | |
|---|---|---|---|---|---|---|---|
| | *DF* | *Blend DF sample + additive* | | | | | |
| | | *A* | *B* | *C* | *D* | *F* | *G* |
| 1 | -4 | -11 | -8 | -17 | -30 | -8 | -26 |
| 2 | -24 | -31 | -33 | -26 | -31 | -32 | -35 |
| 3 | -17 | -21 | -24 | -28 | -27 | -28 | -32 |
| DF sample | $\Delta$ CFPP | | | | | | |
| | *av.* | *A* | *B* | *C* | *D* | *F* | *G* |
| 1 | 12.7 | 7 | 4 | 13 | 26 | 4 | 22 |
| 2 | 7.3 | 7 | 9 | 2 | 7 | 8 | 11 |
| 3 | 9.7 | 4 | 7 | 11 | 10 | 11 | 15 |
| $\Delta$ CFPP av. | | 6.0 | 6.7 | 8.7 | 14.3 | 7.7 | 16.0 |

For samples No. 1, 2, the most reducing of $T_{CP}$ is additive B; on $T_{CP}$ of No. 3 additives B, D and G had an equal effect. On average, the most effective additive for $T_{CP}$ depression is additive B. The Greatest effect of the additive was on $T_{CP}$ of No. 3 DF sample.

If speaking about CFPP depression than the most effective additive for sample No. 1, is additive D; for samples No. 2, 3 the most effective additive is the additive G. On average, the most effective additive for CFPP depression is additive G; the maximum effect from additive using has occurred the sample DF No. 1.

Table 13. The change in $T_{PP}$ of DF samples with adding low-temperature additive

| DF sample | $T_{PP}$,°C | | | | | | |
|---|---|---|---|---|---|---|---|
| | *DF* | *Blend DF sample + additive* | | | | | |
| | | *A* | *B* | *C* | *D* | *F* | *G* |
| 1 | -18 | -30 | -48 | -28 | -25 | -27 | -37 |
| 2 | -45 | -50 | -53 | -50 | -57 | -56 | -53 |
| 3 | -29 | -42 | -48 | -39 | -57 | -54 | -50 |
| DF sample | $\Delta T_{PP}$ | | | | | | |
| | *av.* | *A* | *B* | *C* | *D* | *F* | *G* |
| 1 | 14.5 | 12 | 30 | 10 | 7 | 9 | 19 |
| 2 | 8.2 | 5 | 8 | 5 | 12 | 11 | 8 |
| 3 | 19.3 | 13 | 19 | 10 | 28 | 25 | 21 |
| $\Delta T_{PP}$ av. | | 10.0 | 19.0 | 8.3 | 15.7 | 15.7 | 16.0 |

If speaking about $T_{PP}$ depression than the most effective additive for sample No. 1, is additive B; for samples No. 2, 3 the most effective additive is the additive D. On average, the most effective additive for $T_{PP}$ depression is additive B; the maximum effect from additive using has occurred the sample DF No. 3.

Studies indicate that additives used for research, have a depressors nature as in the fact that their used no effect on $T_{CP}$, but greatly reduced the CFPP and $T_{PP}$. Additives have a significant effect on the low-temperature properties of DF samples No. 1 and 3, and least effect on sample No. 2 properties, due to the effect of DF composition on the effectiveness of low-temperature additives.

### 3.4. Analysis of the influence the DF samples composition on the effectiveness of low-temperature additives

Analyzing data from the study is should be noted following:

The heavier DF (than higher $FC_{IBP}$, $FC_{90\%}$, and density), the more effective low-temperature additives act on $T_{PP}$ and CFPP of fuel (sample No. 1 is the heaviest of the considered – low temperature additives are extremely effective in depressing $T_{PP}$ and CFPP of a given sample, at the same time sample No. 2 is the lightest of the considered, the effectiveness of additives on the low-temperature properties of this sample is one of the lowest);

The higher the total content of n-paraffins in the sample, the lower the effectiveness of the additive on $T_{PP}$ (sample No. 2: the highest content of n-paraffins – the lowest $T_{PP}$ depression, when using the additive);

The higher the content of heavy n-paraffins $C_{20+}$ in the sample, the more effective low-temperature additives act on CFPP depression (sample No 1: the highest content of n-paraffins of $C_{20+}$ – the greatest depression in the CFPP, when using the additive);

The lower the total aromatic hydrocarbon content in the sample, the more effective low-temperature additives act on $T_{PP}$ (sample No. 3: the lowest content of aromatic hydrocarbons – the greatest depression in the $T_{PP}$, when using the additive);

The higher the average number of rings in aromatic hydrocarbon molecules, the more effective low-temperature additives act on $T_{PP}$ (sample No. 3: the largest number of rings in aromatic hydrocarbon molecules – the greatest depression in the $T_{PP}$, when using the additive).

The study results are explained in the mechanism of depressant additives action. The n-paraffins have the highest susceptibility to depressant additives because depressors are designed to interact with the nascent crystals of n-paraffins, thereby stopping their growth.

However, if n-paraffins in the fuel composition will be too much (in the samples under consideration, the content of n-paraffins on average is 20% wt., which is significant), the additive will not cope. However, the depressant additive cannot begin to act until the first crystals of n-paraffins appear, and therefore, the presence of heavy n-paraffins, crystals of which appear the first, increases the effectiveness of depressant additives.

Molecules of aromatic hydrocarbons are more polar than molecules of n-paraffins and can attract the action of a depressant additive on themselves, thereby reducing the effectiveness of the additive on n-paraffins. However, with the increase in the number of aromatic rings in the molecule, the polarity of aromatic hydrocarbons decreases, and the effectiveness of depressant additives by fuel increases.

The established trends explain the highest efficiency of low-temperature additives with samples No. 1 (relatively low total content of n-paraffins, the highest content of heavy n-paraffins, relatively high content of aromatic hydrocarbons, a relatively large number of rings in aromatic molecules) and No. 3 (relatively low total content of n-paraffins, the lowest content of aromatic hydrocarbons, the largest number of rings in aromatic molecules), as well as the lowest efficiency of low-temperature additives with sample No. 2 (the highest total content of n-paraffins, relatively low content of heavy n-paraffins, relatively high content of aromatic hydrocarbons).

## 4. Conclusion

The following results were obtained from a study:

1. The characteristics (density, viscosity, cetane index, cloud point, cold filter plugging point, pour point) of straight-run DF samples was determined. It is established that none of the samples did meet the requirements USS 305-2013 "Diesel fuel. Specifications", applicable to the brands of diesel fuel A and W.
2. The composition (fractional, group, structural-group, sulfur, and n-paraffin content) of straight-run DF samples was determined. It is established that the predominant group of hydrocarbons in the straight-run DF samples composition are paraffins; the highest content of n-paraffins is characteristic of the DF sample No. 2; the highest content of n-paraffins $C_{20+}$ is typical for the DF sample No. 1.
3. The effect of additives on the low-temperature properties of straight-run DF samples is investigated. Found that on average, the most effective additive for $T_{CP}$ depressing is additive B; for CFPP depressing – additive G; for $T_{PP}$ depressing – additive B. It is shown that the additives have a significant influence on low-temperature properties of samples No. 1 and 3, and to a lesser influence on low-temperature properties of sample No. 2, due to the effect of DF composition influence on the effectiveness of low-temperature additives.
4. The influence of the composition of straight-run DF samples on the efficiency of low-temperature additives was investigated. It is shown that with an increase in the total content of n-paraffins in the composition of DF, the effectiveness of low-temperature additives is reduced while with an increase in the content of heavy n-paraffins, the effectiveness of the additives is increased. It is established that with the increase in the total content of aromatic hydrocarbons in the composition of DF, the effectiveness of low-temperature additives decreases, with an increase in the content of polycyclic aromatic hydrocarbons, the effectiveness of the additives increases.

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***List of symbols***

| | | | |
|---|---|---|---|
| *DF* | *diesel fuel;* | $FC_{50\%}$ | *boiling point of 50 %vol. fraction, °C;* |
| *CI* | *cetane index, points;* | $FC_{90\%}$ | *boiling point of 90 %vol. fraction, °C;* |
| $T_{cp}$ | *cloud point,°C;* | $K_{ar}$ | *average number of aromatic rings;* |
| *CFPP* | *cold filter plugging point, °C;* | $K_n$ | *average number of naphthenic rings;* |
| $T_{pp}$ | *pour point, °C;* | $K_t$ | *total average number of rings.* |

| | | | |
|---|---|---|---|
| *S* | *summer brand of diesel fuel;* | $C_{ar}$ | *hydrocarbon content in aromatic rings, % wt.;* |
| *I* | *inter-season brand of diesel fuel;* | $C_n$ | *hydrocarbon content in naphthenic rings, % wt.;* |
| *W* | *winter brand of diesel fuel;* | $C_{ring}$ | *hydrocarbon content in rings structures, % wt.;* |
| *A* | *arctic brand of diesel fuel;* | $C_{alk.chain}$ | *hydrocarbon content in alkyl chains, % wt.;* |
| $FC_{IBP}$ | *initial boiling point, °C;* | *av.* | *average.* |

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