Evaluation of the Feasibility of a Simultaneous Change in the Fractional Composition and the Involvement of Depressant Additives to Obtain Low-Freezing Diesel Fuels

Ilya Bogdanov, Maria Kirgina, Yana Morozova,* and Andrey Altnov

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ABSTRACT: In this paper, the characteristics of a straight-run diesel fuel sample, narrow diesel fractions, and their blends with a depressant additive are tested and analyzed. The efficiency of the depressant additive action on the low-temperature properties of narrow diesel fractions is evaluated. It is shown that the depressant additive is most effective on the pour point of the heavy diesel fraction (300–360 °C) and cold filter plugging point of the light diesel fraction (180–240 °C). The regularities of the effect of narrow diesel fractions on the effectiveness of the depressant additive are established. It is shown that the addition of narrow diesel fractions has practically no effect on the cloud point of the straight-run diesel fuel sample/depressant additive blend. The addition of narrow diesel fractions has a negative effect on the cold filter plugging point of the straight-run diesel fuel sample/depressant additive blend. The addition of the light diesel fraction and middle diesel fraction (240–300 °C) has no significant effect on the pour point of the straight-run diesel fuel sample/depressant additive blend, but the addition of 5 vol % heavy diesel fraction allows lowering the pour point of the blend by 6 °C. The established regularities are explained from the viewpoint of the depressant additive action mechanism. It was found that for a more effective depressant additive action, it is necessary to take into account the content of various narrow fractions in the composition of diesel fuel. It is established that the simultaneous lightening of the fractional composition of diesel fuel (addition of light fractions and/or removal of heavy fractions) and the addition of a depressant additive to obtain low-freezing diesel fuels is impractical. However, the addition of small amounts of heavy diesel fractions simultaneously with depressant additives increases the possibilities for the production of low-freezing grades of diesel fuel as well as expands the feedstock pool of enterprises for the production of fuel. In the future, the identified regularities of the influence of the narrow diesel fractions will allow us to detail the general mechanism of diesel fuel and depressant additive interaction.

1. INTRODUCTION

The production and consumption of diesel fuel are growing every year. Largely, this growth is facilitated by the use of diesel as a fuel for heavy trucks, process equipment, and electric generators. It is important to note the particularly intensive growth in the consumption of the winter and arctic brands of diesel fuel, associated with the development of new territories with a cold climate, primarily the Arctic, as well as the development of the Northern Sea Route.1−5

Despite the active adoption of biodiesel, its use in the capacity of commercial fuel is impossible due to unsatisfactory physical and chemical properties and operational characteristics. As a rule, the proportion of biodiesel involved in the production of commercial fuel usually does not exceed 20 vol %.6,7

The most important and at the same time difficult to achieve characteristics of diesel fuel are its low-temperature properties, which provide the possibility of operating the fuel at negative temperatures. The reason for the unsatisfactory low-temperature properties of diesel fuel is associated with the presence of normal paraffin hydrocarbons in its composition.8

The main methods for obtaining low-freezing diesel fuel are as below9,10

- limiting the content of n-paraffins in the heavy part of the fuel by lowering the end boiling point of the fraction or mixing with lighter fractions (for example, kerosene fractions),
- catalytic processing, such as dewaxing, and
- using the low-temperature (depressant) additives.

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The most effective and profitable way to achieve low-temperature characteristics that meet the standards required is to use depressant additives. In some cases, additives can be combined to obtain the best effect.\textsuperscript{11} However, due to the specific interaction mechanism of depressant additives with hydrocarbons that make up diesel fuel, it is possible that the additives do not make the expected improvement in low-temperature characteristics.\textsuperscript{11–13} Also, the opposite situation is possible when the presence of certain hydrocarbons in the composition of diesel fuel intensifies the effect of the additive.\textsuperscript{15}

The influence of the diesel fuel’s group hydrocarbon composition on the effectiveness of depressant additives with various structures was studied in refs 13, 16, 17. But not just the content of various groups of hydrocarbons influences the performance properties of fuels; in particular, the ratio of high- and low-melting-point \(n\)-alkanes and isoalkanes also influences the fuel properties.\textsuperscript{18} However, based on the data obtained, it was found that \(n\)-paraffins are the most susceptible to depressant additives, which is due to the mechanism of their action.\textsuperscript{14,19} However, \(n\)-paraffins themselves are characterized by close to positive pour points, which worsens the low-temperature properties of fuels. In this connection, the question arises about the optimal concentration of \(n\)-paraffins in the fuel.

In the context of large-scale production, the adjustment of the individual hydrocarbon composition of diesel fuel is practically impossible; however, it is possible to adjust the fractional composition by increasing or decreasing the proportion of various components involved in the blending of commercial fuel. A change in the fractional composition of diesel fuel can also affect the efficiency of the depressants, since, depending on the boiling range, the fractions contain certain groups of hydrocarbons. This work studies the effect of the different content of narrow diesel fractions on the effectiveness of depressant additives. The aim of this work is to evaluate the feasibility of simultaneous change in the fractional composition and involvement of depressant additives for upgrading the low-temperature properties of diesel fuel.

2. MATERIALS AND METHODS

The objects of the research are the straight-run diesel fuel (DF) sample, narrow diesel fractions (NDFs) as well as their blends, and the blends with depressant additives. The samples used in this study were obtained with an industrial atmospheric oil distillation unit located at an oil field in Western Siberia, Russian Federation.

NDFs were separated from the atmospheric diesel fraction with the boiling range of 161–359 °C and density at 15 °C—871.9 kg/m\(^3\). The separation of NDFs was implemented using a distillation unit. The following NDFs were separated:

- light diesel fraction (LDF): 180–240 °C boiling range,
- middle diesel fraction (MDF): 240–300 °C boiling range, and
- heavy diesel fraction (HDF): 300–360 °C boiling range.

For the study, blends of a straight-run DF sample with each of the NDF were prepared, the contents of which in the blend were 1, 3, 5, and 10 vol %, as well as similar blends with the addition of a depressant additive (Ad). The depressant additive was used in the concentration recommended by the manufacturer—0.3 mL of the additive per 100 mL of DF.

To determine the physicochemical and operational characteristics of the straight-run DF sample, NDFs, as well as their blends, the following methods were used:

- The fractional composition was determined according to ISO 3405:2011 "Petroleum products—Determination of distillation characteristics at atmospheric pressure"\textsuperscript{20}
- The density at a temperature of 15 °C was determined using a Stanbinger SVM3000 Anton Paar viscometer according to ISO 12185:1996 “Crude petroleum and petroleum products—Determination of density—Oscillating U-tube method”\textsuperscript{21}
- The kinematic viscosity at 20 °C was determined using the Stanbinger SVM3000 Viscometer Anton Paar, according to ISO 3104:1994 "Petroleum products. Transparent and opaque liquids. Determination of kinematic viscosity and calculation of dynamic viscosity"\textsuperscript{22}
- The sulfur content was determined using X-ray fluorescence energy dispersive analyzer “SPECTROSCAN S”, according to ASTM D4294-16 “Standard Test Method for Sulfur in Petroleum and Petroleum Products by Energy Dispersive X-ray Fluorescence Spectrometry”\textsuperscript{23}
- The cloud point (CP) was determined using the liquid low-temperature thermostat CRYO-T-05-01 according to ASTM D2500-05 “Standard Test Method for Cloud Point of Petroleum Products”.\textsuperscript{24}
- The cold filter plugging point (CFPP) was determined using the liquid low-temperature thermostat CRYO-T-05-01 and the cold filter plugging point measuring unit according to ASTM D6371-17a “Standard Test Method for Cold Filter Plugging Point of Diesel and Heating Fuels”.\textsuperscript{25}
- The pour point (PP) was determined using the liquid low-temperature thermostat CRYO-T-05-01 according to ASTM D97-17b “Standard Test Method for Pour Point of Petroleum Products”.\textsuperscript{26}
- The cetane index was determined according to ISO 4264:2018 “Petroleum products—Calculation of cetane index of midle-distillate fuels by the four variable equation”, and it is the most accurate calculation method.\textsuperscript{27}
- To determine the content of \(n\)-paraffins in the NDFs, the method of gas—liquid chromatography was applied (Chromatec-Crystal 5000 chromatograph with a quartz capillary column 25 m × 0.22 mm, stationary phase—SE-54, carrier gas—helium).

3. RESULTS AND DISCUSSION

3.1. Physicochemical and Operational Characteristics of the Straight-Run DF Sample. The results of the determination of physicochemical and operational characteristics of the straight-run DF sample are presented in Table 1. The results of various physicochemical and operational characteristics of the straight-run DF sample were compared with the Interstate standard of Armenia, Kyrgyzstan, Russia, and Uzbekistan\textsuperscript{29} for diesel fuels. As can be seen from Table 1, the straight-run DF sample meets the requirements of IS 305-2013 in terms of density (no more than 843.4 kg/m\(^3\)) and kinematic viscosity (1.8–5.0 mm\(^2\)/s) for summer, interseason, and winter grades of DF. In terms of sulfur content, the straight-run DF sample does not meet the requirements of IS 305-2013 (no more than 2000 mg/kg). To use the investigated straight-run DF sample as a commercial fuel, its preliminary desulfurization is required.
As can be seen from the data presented in Table 3, LDF has the best low-temperature properties and H_{DF} has the worst. It should be noted that the CP and CFPP of light and medium NDFs are negative; at the same time, these characteristics of the diesel fraction, used for separating NDFs, are positive. These results are explained by the composition of the fractions—L_{DF} and M_{DF} practically do not contain heavy n-paraffins, the presence of which significantly impairs the low-temperature properties of the fuel.

Based on the obtained results, it can be concluded that the addition of an additive to the studied straight-run DF sample practically does not change its CP (decrease only by 2 °C); however, it has a significant effect on CFPP and PP—each of the characteristics decreased by 20 °C. A straight-run DF sample with the addition of a depressant meets the requirements of IS 305-2013 in terms of CFPP (not higher than −25 °C) for a winter grade of fuel.

The addition of a depressant additive to NDFs has a positive effect on their low-temperature properties. Figure 1 shows the change in the low-temperature properties of NDFs upon the addition of a depressant to them.

As can be seen from Figure 1, the additive has a different effect on the fractions with different boiling ranges. The addition of a depressant is most effective on PP of H_{DF}. The revealed effect is explained by the mechanism of the depressant additive action. The content of n-paraffins in H_{DF} is higher than in the middle and light fractions; therefore, crystal formation occurs faster, which entails a faster onset of the additive’s action and, as a result, leads to a greater effect.

In terms of CFPP, the addition of a depressant is the most effective for L_{DF}; however, the effectiveness in terms of PP is the lowest. This effect is connected with the size of the crystals. The n-paraffin crystals formed in the light fraction are smaller than the crystals contained in the medium and heavy fractions, which makes it much easier for them to pass through a standard filter element. The depressant additionally prevents small crystals uniting into larger structures.

The results of the study showed that the CP of M_{DF} is more susceptible to the addition of a depressant than the CP of L_{DF} and H_{DF}. This effect is presumably connected with the highest content of monocyclic aromatic hydrocarbons in M_{DF}, which are the solvents for n-paraffins. The presence of a depressant in the blend prevents the growth of small crystals, which is manifested in a decrease of the CP.

The obtained results mean that for a more effective depressant additive action, it is necessary to take into account the content of...
Various narrow fractions in the DF since, depending on their content, the effectiveness of the additive will change.

**3.4. Low-Temperature Properties of the Straight-Run DF Sample/NDF/Depressant Additive Blends.** The results of the determination of low-temperature properties of the straight-run DF sample/NDF/depessant additive blends are presented in Table 5.

Table 5. Low-Temperature Properties of the Straight-Run DF Sample/NDF/Depressant Additive Blends

| Blend | Temperature Properties (°C) | CP | CFFP | PP |
|-------|-----------------------------|----|------|----|
| DF + 1% LDF + Ad | -6 | -20 | -37 |
| DF + 3% LDF + Ad | -5 | -15 | -36 |
| DF + 5% LDF + Ad | -6 | -20 | -35 |
| DF + 10% LDF + Ad | -6 | -19 | -36 |
| DF + 1% MDF + Ad | -6 | -17 | -34 |
| DF + 3% MDF + Ad | -7 | -21 | -36 |
| DF + 10% MDF + Ad | -7 | -20 | -35 |
| DF + 1% HDF + Ad | -5 | -19 | -34 |
| DF + 3% HDF + Ad | -6 | -19 | -35 |
| DF + 5% HDF + Ad | -6 | -17 | -42 |
| DF + 10% HDF + Ad | -6 | -17 | -36 |

As can be seen from Figure 2, the addition of LDF into the straight-run DF/depessant additive blend leads to a decrease of the additive effect on CFFP. The greatest negative effect is observed for the blend with the addition of 3 vol % LDF. The reduction of the negative effect on CFFP with the addition of higher concentrations of LDF (5, 10 vol %) connected with a general improvement in the low-temperature properties of the blend. The addition of LDF, which is characterized by good low-temperature properties, compensates the negative effect of LDF on the effectiveness of the additive. At the same time, it can be seen that the addition of LDF has practically no effect on CP and PP for all blends (temperature variation by ±1 °C is within the error limits of experimental methods for determining these properties). The obtained results are consistent with the concept of the action mechanism of the depressants. The addition of the diesel fraction with 180–240 °C boiling range reduces the content of the heaviest (high-boiling, readily solidifying) hydrocarbons in the blend, which quickly crystallize. Depressant additives can begin to act only when the first crystals of n-paraffins are formed, but in this case, CFFP with the addition of LDF deteriorates, since the additive does not have time to interact with all of the formed small crystals of light n-paraffins, but their amount is sufficient to plug a standard filter element.

Thus, it can be concluded that the simultaneous addition of LDF and the depressant additive to improve the low-temperature properties of DF is inappropriate. The addition of a LDF not only does not increase the effectiveness of the depressant additive but also has a negative effect on CFFP in comparison with a blend only with an additive and without LDF added.

**3.5. Investigation Results of the Influence of MDF Addition on the Effectiveness of the Depressant Additive Action.** Figure 2 shows the change in the low-temperature properties of the straight-run DF sample/depessant additive blend upon the addition of different MDF concentrations to it.

As can be seen from Figure 3, the addition of MDF into the straight-run DF sample/depessant additive blend leads to a decrease of the additive effect on CFFP, but with an increase in the content of MDF in the blend, the negative effect on CFFP
decreases. It can also be seen that the addition of MDF has no effect on CP and PP for all blends (temperature variation by ±2 °C is within the error limits of experimental methods for determining these properties).

The obtained results also find an explanation in the mechanism of depressants’ action. MDF contains light hydrocarbons and relatively heavy ones. The addition of the diesel fraction with 240−300 °C boiling range reduces the content of the heaviest hydrocarbons in the blend, which trigger the action of the depressant additive, and the effectiveness of the additive decreases. At the same time, the addition of MDF increases the content of relatively heavy hydrocarbons (added with MDF) in the blend, which form the first crystallization centers, allowing the depressant to start acting, and the negative effect on CFPP decreases.

Thus, the simultaneous addition of MDF and a depressant additive to improve the low-temperature properties of DF is impractical. A decrease in the negative effect on CFPP can be achieved by adding a significant amount of MDF.

3.7. Investigation Results of the HDF Addition Influence on the Effectiveness of the Depressant Additive Action. Figure 4 shows the change in the low-temperature properties of the straight-run DF sample/
depressant additive blend upon the addition of different H_{DF} concentrations to it.

As can be seen from Figure 4, the addition of H_{DF} into the straight-run DF sample/depressant additive blend leads to a decrease of the additive effect on CFPP; increasing the content of H_{DF} in the blend increases the negative effect on CFPP. It can also be seen that the addition of H_{DF} has no effect on CP for all blends. At the same time, the addition of a small amount of H_{DF} has an insignificant negative effect on the PP of the blend (1–2 °C), but the addition of 5 vol % H_{DF} allows decreasing the PP of the blend by 6 °C, and the addition of 10 vol % H_{DF} does not change the PP of the blend.

The obtained results are also consistent with the mechanism of depressant additive action. The additive begins to act upon the formation of the first paraffin crystals. The addition of the diesel fraction with 300–360 °C boiling range increases the content of the heaviest hydrocarbons, which quickly crystallize and trigger the action of the additive. In this case, the negative effect on CFPP is connected with the size of the heavy hydrocarbon crystals. Despite the fact that the additive stops crystal growth, the original crystals are large enough and, in any case, plug the standard filter element. At the same time, the additive still prevents the crystals from associating into a full-fledged solid structure, which is confirmed by the results obtained for PP.

Thus, it can be concluded that the simultaneous addition of the depressant additive and small amounts of H_{DF} to improve the low-temperature properties of the DF is expedient. It should be noted that the results obtained are of great practical importance since the involvement of H_{DF} in the production of commercial fuels allows to significantly increase the feedstock pool of enterprises.

4. CONCLUSIONS

(1) It is established that the straight-run DF sample in terms of low-temperature properties meets the requirements for the summer grade of commercial DF. It is shown that the use of the investigated straight-run DF sample in winter or arctic conditions is possible only after upgrading or in the case of using depressant additives. It was found that for a more effective depressant additive action, it is necessary to take into account the content of various narrow fractions in the composition of DF. Also, it is shown that all straight-run DF sample/depressant additive/NDF blends meet the requirements for the interseason grade of commercial DF in terms of low-temperature properties.

(2) The regularities of the effect of NDFs on the effectiveness of the depressant additive are established. It is shown that the addition of NDFs has practically no effect on the CP of the straight-run DF sample/depressant additive blend. On the CFPP of the straight-run DF sample/depressant additive blend, the addition of NDFs has a negative effect. With an increase in the content of L_{DF} and M_{DF} in the blend, the negative effect decreases, but with an increase in the content of H_{DF} in the blend, the negative effect increases. The greatest deterioration of CFPP (increase by 10 °C) is observed with the addition of 3 vol % of L_{DF}. The addition of L_{DF} and M_{DF} has no significant effect on PP of the straight-run DF sample/depressant additive blend (a change is observed within the error limits of experimental methods for determining this property), but the addition of 5 vol % H_{DF} allows lowering PP by 6 °C. Thus, it is shown that to improve the PP of DF, it is advisable to involve small amounts of H_{DF} simultaneously with a depressant additive; the involvement of M_{DF} and L_{DF} is impractical, and the involvement of L_{DF} leads to a significant deterioration of the DF CFPP.

(3) It is established that the simultaneous lightening of the fractional composition of DF (addition of light fractions and/or removal of heavy fractions) and the addition of a depressant additive to obtain low-freezing DF are impractical. However, the addition of small amounts of heavy diesel fractions simultaneously with depressant additives increases the possibilities for the production of low-freezing grades of DF as well as expands the feedstock pool of enterprises for the production of DF by involving H_{DF}. In addition, the identified regularities will allow manufacturers of low-temperature additives to create new, more effective, and multifunctional compositions of additives. The identified regularities of the influence of the narrow diesel fractions will allow us to detail the general interaction mechanism of diesel fuel and depressant additives. In the future, studies will be carried out in the area of the influence of individual hydrocarbon content in the composition of diesel fuel using model blends as an example.

Author Information

Corresponding Author
Yana Morozova — School of Earth Sciences & Engineering, Tomsk Polytechnic University, Tomsk 634050, Russia; orcid.org/0000-0002-0531-6405; Email: yana_morozova@tpu.ru

Authors
Ilya Bogdanov — School of Earth Sciences & Engineering, Tomsk Polytechnic University, Tomsk 634050, Russia
Maria Kirgina — School of Earth Sciences & Engineering, Tomsk Polytechnic University, Tomsk 634050, Russia
Andrey Altnynov — School of Earth Sciences & Engineering, Tomsk Polytechnic University, Tomsk 634050, Russia

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.1c06472

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Nomenclature

C_{10}−C_{15} n-paraffin with 10–15 carbon atoms in a molecule
C_{16}−C_{25} n-paraffin with 16–20 carbon atoms in a molecule
C_{26}−C_{30} n-paraffin with 21–25 carbon atoms in a molecule
C_{31+} n-paraffin with more than 31 carbon atoms in a molecule

Abbreviations Used

DF straight-run diesel fuel sample
NDFs narrow diesel fractions
L_{DF} light diesel fraction
M_{DF} middle diesel fraction
HDF  heavy diesel fraction
Ad  depressant additive
CP  cloud point
CFPP  cold filter plugging point
PP  pour point

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