RESEARCH INTO EMISSIONS OF NITROGEN OXIDES WHEN CONVERTING THE DIESEL ENGINES TO ALTERNATIVE FUELS

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1. Introduction

Oil reserves in the depths of the Earth constantly reduce. According to the optimistic forecasts, at existing explored reserves and current levels of oil extraction, petroleum will last for about fifty years. The second energy resource after oil that can be utilized as fuel for engines is natural gas. Many countries of the world have long come to understand how serious and necessary it is to develop alternative energy generation. At present, against the background of different
gas prices and volatile oil prices, there is a constantly rising demand for energy resources, which necessitates the search for innovative solutions to energy problems. Many of the leading countries of the world are actively engaged in searching for alternative energy sources and the implementation of new technologies to increase the volumes of energy resources [1].

Currently, natural gas is used in most countries of the world as a motor fuel. Low price and high environmental characteristics contribute to utilizing gas as a motor fuel [2]. Studies show that diesel engines, which are converted to alternative fuels, have very high traction, dynamic and economic characteristics, and even significantly outperform basic diesel engines in terms of environmental impact.

This allows us to assert that in order to solve a comprehensive global task on reducing the concentration rate of liquid petroleum fuels, it is necessary to significantly increase the number of power drives with engines that run on alternative fuels. To this end, among other areas, it is required to create for engines of motor transport and other machines a technology for converting existing diesel power drives into gas engines. In this case, the diesel power drives that would be converted into such gas engines will make it possible to reduce the negative impact of engines on the environment.

One of the most adverse environmental problems at the present stage of development of designs of internal combustion engines is the emission of nitrogen oxides in exhaust gases. This issue is especially relevant for modern diesel engines that operate on very lean fuel mixtures. Thus, reducing the levels of emissions of nitrogen oxides in diesel engines by their conversion to gas fuel is an important task for specialists in the field of design and operation of internal combustion engines.

### 2. Literature review and problem statement

When operating internal combustion engines, the oxides of nitrogen form as a result of the oxidation of nitrogen by air oxygen in the process of fuel combustion. The processes of formation of nitrogen oxides during combustion of various fuels were investigated in paper [3]. The authors established that the concentration of nitrogen oxides in the exhaust gases is affected by both nitrogen from the air and the nitrogen contained in fuel; however, effect of nitrogen in the fuel on the formation of nitrogen oxides was overstated.

Paper [4] investigated quantitative characteristics of nitrogen oxides formation during fuel combustion. It was established that the amount of nitrogen oxides depends on the type of engine, parameters of the working process and the environment; however, no effect of operation modes of an engine on the formation of nitrogen oxides was examined.

Study into concentration of nitrogen oxides in exhaust gases of the diesel engines that run on a mixture of methanol with diesel fuel was reported in papers [5, 6]. It was found that one of the main factors that affect the emissions of nitrogen oxides is the temperature of combustion in the combustion chamber [5]. It was established in [6] that the main amount of nitrogen oxides is produced in diesel engines during the first phase of combustion. The specified papers, however, failed to investigate influence of the angle of rotation of the engine crankshaft on the concentration of nitrogen oxides in exhaust gases.

Interesting research into reduction of ecocarcinogenic danger of vehicles through the use of alternative fuels was undertaken in paper [7]. It is shown that the formation of 90 % of carcinogenic substances contained in exhaust gases is due to the process of combustion. The task, resolved by the author of the work, implies the establishment of an integrated index of ecocarcinogenic danger of automobiles, though the criteria were not defined for its compliance with current international standards taking into account sanitary-hygienic regulations for carcinogenic and toxic components.

The automotive corporation KamAZ converted the diesel engine KamAZ-740 into a gas engine operating on methane in line with the environmental standards Euro 5 with a variable feed of air-gas mixture to the engine inlet system and spark ignition [8]. The engineers performed laboratory and field tests of the designed engine and found that the converted gas engine brought down the emissions indicators for nitrogen oxides by 35 % compared to the basic diesel engine. However, the author paid no attention to the link between the emission of nitrogen oxides and the capacity taken from the engine.

Authors of paper [9] performed bench tests of the diesel engine D21A1 when operating on mixtures of diesel fuel and fuel oils. The experiments revealed that an increase in the share of fuel oils in fuel mixtures led to an increase in the content of nitrogen oxides by 5–15 %. The problem reported is in the fact that the use of some alternative fuels leads to lower fuel costs but, at the same time, increases emissions of carcinogenic substances.

Study into using alcohols as a fuel for the diesel engines is described in [10]. Based on the diesel engine 1Ch 12/14, authors designed an experimental setup and found that using 50 % of methyl alcohol in the mixture reduced emissions of nitrogen oxides by 30 %. However, it was determined whether the emissions of nitrogen oxides are affected by rotation frequency of the engine crankshaft.

Development and estimation of particulate filters for the process of continuous regeneration of nitrogen oxides were reported in paper [11]. The article described a constructed mathematical model of nitrogen oxides regeneration and the loss of pressure in the filters of nitrogen oxides regeneration. The authors did not experimentally verify the proposed mathematical model.

Study [12] explored ways of reducing harmful emissions from automobiles into atmosphere and techniques for converting automobile engines to alternative fuels. The main direction of research was the development of methods to inhibit the rate of global warming. However, the authors did not propose any adequate mathematical model to describe the mechanisms of formation of excess heat.

Much attention is paid to the emissions of nitrogen oxides, soot and carbonyl compounds from diesel and gasoline engines of passenger cars based on the standards Euro 4 and Euro 5 [13]. Tests were conducted for four vehicles in line with Euro 5 and two cars in line with Euro 4 with diesel engines, equipped with the additive and catalytic filters. The results showed that in comparison with cycles of hot start the cycles of cold start increased the emissions of all pollutants by up to two times. The authors, however, paid insufficient attention to checking the results at a dynamic bench.

Statistical data on 39 diesel passenger buses that comply with standards Euro 6 on a test route were gathered by American specialists [14]. The results revealed a large
difference in emissions of nitrogen oxides on urban routes depending on the brand of the vehicle. The average emissions of nitrogen oxides on urban routes exceeded the established norms of Euro 6 by 4.5 times. The authors, however, failed to establish links between the emissions of nitrogen oxides and rotation frequencies of engine crankshafts on city routes.

Thus, at present, there is a need to determine the content of toxic components in the exhaust gases of diesel engines, re-equipped for gas fuel, including the content of nitrogen oxides $r_{NO}$, given the absence of adequate mathematical models that would make it possible.

3. The aim and objectives of the study

The aim of present study is to examine the emissions of nitrogen oxides in the exhaust gases of diesel engines and in engines re-equipped for gas fuel. This is necessary to establish actual levels of emissions of carcinogenic substances from diesel engines re-equipped for gas fuel. In practice, this would make it possible not only to reduce fuel costs, but significantly lower emissions of toxic components from the converted diesel engines.

To accomplish the aim, the following tasks have been set:

– to build a mathematical model for estimating the emissions of nitrogen oxides in the exhaust gases of diesel engines and engines re-equipped for gas-propane-butane mixture;

– to establish patterns of change in the emissions of nitrogen oxides when operating on diesel fuel and when operating on gas fuel at stable rotation frequency of the crankshaft depending on a change in loading;

– to establish patterns of change in the emissions of nitrogen oxides in the exhaust gases of an engine when operating on diesel fuel and when operating on gas fuel, depending on rotation frequency of the engine crankshaft.

4. Construction of a mathematical model for estimating the emissions of nitrogen oxides in exhaust gases

The mathematical model to be constructed will make it possible to perform calculations of the emissions of nitrogen oxides in the exhaust gases of diesel engines re-equipped for gas fuel under different modes of operation.

During combustion of gas fuel in a diesel engine, re-equipped for gas, nitrogen oxides form as a result of reaction of nitrogen oxidation with oxygen in the air. In this case, oxidation reaction mainly involves nitrogen from the atmospheric air, but in the presence of nitrogen in fuel, the specified nitrogen will also participate in the process of formation of nitrogen oxides during combustion.

The calculation procedure will be based on the basic principles of heat theory:

– the amount of nitrogen oxides is determined by the maximum temperature of combustion and the concentrations of nitrogen and oxygen in the combustion products;

– the amount of nitrogen oxides depends on the cooling rate of combustion products;

– formation of nitrogen oxides occurs more intensively under conditions of a leaner air-fuel mixture;

– nitrogen oxidation occurs as a result of the chain mechanism and in the zone of combustion products.

The following assumptions are accepted for calculations:

– the amount of nitrogen oxides is not dependent on the chemical nature of fuel; temperatures and pressures in the zone of combustion products are the same at all points in the combustion chamber at a given moment of the engine cycle; combustion products are homogeneous in their composition; the charge of the cylinder is known, as well as the fuel combustion law and the indicator chart.

In general, the mechanism of nitrogen oxides formation consists of the following reactions:

$$N + O_2 = NO + O + 136 \text{ kJ/mole}, \quad (1)$$
$$N_2 + O = NO + N - 316 \text{ kJ/mole}, \quad (2)$$
$$N_2 + H_2O = 2NO + H_2, \quad (3)$$
$$N_2 + O_2 = N_2O + O, \quad (4)$$
$$N_2O + O = 2NO. \quad (5)$$

Basic oxides formed in the exhaust gases of internal combustion engines are NO and N_2O. Based on equations (1)–(5), we shall find the rate of formation of NO and N_2O. From the following kinetic equations:

$$\frac{\partial r_{NO}}{\partial \tau} = K'_r \cdot r_{N_2} \cdot r_{O} - K''_r \cdot r_{NO} \cdot r_{O} +$$

$$+ K'_r \cdot r_{O} \cdot r_{N_2O} - K''_r \cdot r_{NO} \cdot r_{N_2O} +$$

Then the volumetric concentration of diazate oxide, in line with (7), will equal

$$r_{N_2O} = K'_r \cdot r_{N_2} \cdot r_{O} - K''_r \cdot r_{NO} \cdot r_{O} +$$

$$+ K'_r \cdot r_{O} \cdot r_{N_2O} - K''_r \cdot r_{NO} \cdot r_{N_2O}.$$  

By substituting (8) in (6), we shall obtain

$$\frac{\partial r_{NO}}{\partial \tau} = K'_r \cdot r_{N_2} \cdot r_{O} - K''_r \cdot r_{NO} \cdot r_{O} +$$

$$- K'_r \cdot r_{N_2OT} + K'_r \cdot r_{N_2O} \cdot r_{O} - K''_r \cdot r_{NO} \cdot r_{N_2O} +$$

$$+ K'_r \cdot r_{O} \cdot K'_r \cdot r_{N_2O} - K''_r \cdot r_{NO} \cdot r_{N_2O}.$$  

To convert rate constants from cm$^3$ molecules$^{-1}$ to (%·s)$^{-1}$, we shall use equation

$$\frac{\partial r_{NO}}{\partial \tau} = 0.$$
\[ K' = \frac{K' \cdot p}{8.314 \cdot 10^8 T}. \]

where \( p \) is the pressure of working body in the combustion chamber, Pa.

Dependences of rate constants of chemical reactions \( K'_{ni} \), \( K'_{ni} \) on the temperature of combustion of air-fuel mixtures \( T \) are taken based on an analysis of the scientific literature [15–19] and are given in Table 1.

| Rate constants of chemical reactions | Constant, cm\(^3\) molecule\(^{-1}\) s\(^{-1}\) | Source |
|-------------------------------------|---------------------------------|--------|
| \( K'_{ni} = 1.337 \cdot 10^{10} e^{-\frac{153800}{T}} \), | (10) | [15] |
| \( K'_{ni} = 3.2 \cdot 10^3 \cdot 10^{\frac{107380}{T}} \), | (11) | |
| \( K'_{ni} = 1.36 \cdot 10^{11} \cdot e^{-\frac{157280}{T}} \), | (12) | [16] |
| \( K'_{ni} = 3.12 \cdot 10^{17} \cdot e^{-\frac{20720}{T}} \), | (13) | |
| \( K'_{ni} = 4.2 \cdot 10^3 \), | (14) | [17] |
| \( K'_{ni} = 1.3 \cdot 10^{11} \cdot e^{-\frac{22900}{T}} \), | (15) | |
| \( K'_{ni} = 1.0 \cdot 10^{17} \cdot e^{-\frac{10400}{T}} \), | (16) | [18] |
| \( K'_{ni} = 8.59 \cdot 10^9 \cdot e^{-\frac{32340}{T}} \), | (17) | |
| \( K'_{ni} = 1.52 \cdot 10^7 \cdot e^{-\frac{15980}{T}} \), | (18) | [19] |
| \( K'_{ni} = 1.0 \cdot 10^7 \cdot e^{-\frac{14800}{T}} \), | (19) | |

We shall determine the rate of nitrogen oxides formation based on the angle of rotation of the crankshaft from a differential equation of nitrogen oxides formation on time (9) with respect to \( \tau = \frac{\phi}{60} \). Dependence of the rate of nitrogen oxides formation on the angle of rotation of the crankshaft will be written in the form of differential equation

\[ \frac{d n_{SO}}{d \phi} = A + B \cdot r_{SO} + C \cdot r_{SO}^2, \]

where \( A, B, C \) are the initial constants that correspond to the calculation step, \( \Delta \phi \) is the calculation interval step, ° p.c.s.

Step of interval \( \Delta \phi \) at a given sampling frequency \( N \) and the frequency of rotation of the crankshaft \( n \) is determined with respect to \( \Delta \phi = \frac{\pi r}{60} \). By integrating differential equation (20) relative to \( \phi \), we shall obtain:

\[ r_{SO} = r_{SO0} e^{\left(\frac{B}{2C} + \beta \frac{B}{2C} \cdot \phi \cdot \frac{B}{2C}ight)} \cdot \left( e^{\left(\frac{B}{2C} + \beta \frac{B}{2C} \cdot \phi \cdot \frac{B}{2C}ight)} - 1 \right), \]

where \( \Delta \phi = \frac{\pi r}{60} \).

Denote \( \beta = \sqrt{\frac{B^2 - 4AC}{4C^2}} \). Because, according to expressions (21) and (23), constants \( A \geq 0 \) and \( C < 0 \), the value \( \beta \) always exists and \( \beta \geq 0 \). Then the volumetric concentration of nitrogen oxides can be determined from formula in the form:

\[ r_{SO} = r_{SO0} e^{\left(\frac{\pi r}{60} \frac{B}{2C} \cdot \phi \cdot \frac{B}{2C}ight)} \cdot \left( e^{\left(\frac{\pi r}{60} \frac{B}{2C} \cdot \phi \cdot \frac{B}{2C}ight)} - 1 \right). \]

In expression (25), if \( r_{SO} > \frac{B}{2C} \cdot \frac{B}{2C} \cdot \frac{B}{2C} \), we accept the plus sign, if \( r_{SO} < \frac{B}{2C} \cdot \frac{B}{2C} \cdot \frac{B}{2C} \), we accept the minus sign.

Therefore, expression (25) describes the mathematical model which makes it possible to determine the starting concentration of nitrogen oxides \( r_{SO0} \) in the exhaust gases from diesel engines, re-equipped for gas fuel, depending on the original parameters: rate constants of chemical reactions \( K'_{ni} \), \( K'_{ni} \), starting concentration of nitrogen oxides \( r_{SO0} \), change in angle of rotation \( \phi \) of the engine crankshaft and the temperature of combustion of an air-gas mixture \( T \). A range of change in the temperature of combustion of fuel-air mixtures \( T \) varies within 1,800–2,400 K, rotation angle \( \phi \) of the crankshaft changed in the range of 0–720°.

5. Methods and materials of experimental research into emissions of nitrogen oxides

The purpose of experimental research is to test the developed mathematical model for calculating the emissions of nitrogen oxides in the exhaust gases of diesel engines, re-equipped for gas fuel. To perform the test task, specialist at Ivan Frankivsk National Technical University of Oil and Gas (Ukraine) re-equipped for gas fuel the diesel engine X17DTL of the car Opel Astra (Fig. 1).

The specified car and its engine were manufactured in 2000 and by the time the research started the car had traveled 186 thousand km. Compression in the engine cylinders was \( 2.95 \pm 0.05 \) MPa. To re-equip the vehicle’s diesel engine X17DTL for the model Opel Astra for gas, the degree of engine compression was reduced and propane-butane gas equipment, made in Italy, was installed. Then we added the original microprocessor DIS-ignition system of our own design and optimized engine control system operation.
Fig. 1. Diesel engine of Opel Astra make re-equipped for a propane-butane mixture: a — diesel engine of model X17DTL; b — converted engine cylinder head; c — gas cylinder equipment

Brief technical specifications of the re-equipped diesel engine X17DTL, Opel Astra make, are given in Table 2.

| No. of entry | Parameter name                      | Specification                                                                 |
|--------------|-------------------------------------|-------------------------------------------------------------------------------|
| 1            | Base engine                         | Diesel, Bosch electronic injection system                                    |
| 2            | Converted engine                    | Gas (propane-butane mixture) with electronic operation system, designed at IFNTUOG |
| 3            | Working volume of engine, cm³       | 1,669                                                                         |
| 4            | Rated power, kW (h.p.)              | 50 (68)                                                                       |
| 5            | Crankshaft rotation frequency at operational power, rpm | 4,500                                                                         |
| 6            | Base engine compression pressure     | 22.0                                                                          |
| 7            | Compression of engine converted to gas | 13.1                                                                         |

Table 2

The motor study was performed at the electric brake bench KI-8964 GOSNITI with the range of change in brake force of 1.0–16.0 kN. The bench consists of the following main components: left drum unit, right drum unit, control unit, remote control, pedometer, pedal switch. Physical appearance of the bench KI-8964 GOSNITI and procedure of experimental research are shown in Fig. 2.

Fuel tank of the car was filled with diesel fuel of brand L produced at Kremenchug oil refinery. Nozzle opening pressure was 13.5–13.9 MPa. Volumetric fractions of nitrogen oxides were measured by the gas analyzer “Avtotest-02.03P” (Russian Federation). Measurement range of nitrogen oxides is 0–5,000 ppm, absolute error of measurement is ±10 ppm. When determining the temperature of exhaust gases, we used thermocouples of L type and the logometer-potentiometer UP-2M (Ukraine). In order to perform comparative estimation of environmental indicators of engine re-equipped for gas against respective indicators of the diesel engine, we registered loading characteristics of the engine at fixed rotation frequency of the crankshaft.

6. Results of research into emissions of nitrogen oxides in exhaust gases of the diesel engines, converted to gas fuel

Fig. 3 shows dependences of the emission of nitrogen oxides in exhaust gases of the diesel engine X17DTL, Opel Astra make, when operating on diesel fuel and when operating on propane-butane at rotation frequency of 1,800 rpm dependent on a change in loading.

Based on the results of experiments, it is possible to note that the use of gas fuel, compared to the diesel fuel, led to a significant reduction in the emissions of nitrogen oxides in exhaust gases over the entire loading mode. Thus, when operating on diesel fuel, at a power take-off of 10 kW, the emissions of nitrogen oxides in exhaust gases of the base engine X17DTL, Opel Astra make, amounted to 255 ppm; and when the converted engine operated on a propane-butane mixture – 135 ppm. In other words, the reduction of emissions of nitrogen oxides in the exhaust gases reached 47.1 %. When operating on diesel fuel, at a power take-off of 50 kW, the emissions of nitrogen oxides in exhaust gases of the base engine 2L amounted to 945 ppm, and when the converted engine operated on a propane-butane mixture – 575 ppm. In other words, the reduction of emissions of nitrogen oxides in the exhaust gases reached 39.2 %.
mixture – 570 ppm. In other words, the reduction of emissions of nitrogen oxides in the exhaust gases was 39.7%.

Fig. 4 shows dependences of the emission of nitrogen oxides in exhaust gases of the diesel engine X17DTL, Opel Astra make, when operating on diesel fuel and gas fuel, on the crankshaft rotation frequency. Thus, when the base engine operates at a crankshaft rotation frequency of 1,000 rpm, there is a relatively insignificant reduction in the emissions of nitrogen oxides from 790 ppm for diesel fuel to 680 ppm for gas fuel, or by 13.9%. However, when the base engine operates at a high frequency of 4,000 rpm, there is a significant reduction in the emissions of nitrogen oxides from 655 ppm for diesel fuel to 475 ppm for gas fuel, or by 27.5%.

The mathematical model was checked for adequacy in the following order. We derived theoretical curves and oscillograms of emissions. The obtained theoretical curves and oscillograms are described by the polynomial models, and the character of change in the experimental oscillograms and theoretical curves for the emissions of nitrogen oxides is almost the same. Next, in line with the plan for a symmetrical full-factorial experiment, we determined values of parameters at points in the plan, based on which the coefficients for polynomial models were determined by the least square method. Then, employing values on which the coefficients for polynomial models were determined values of parameters at points in the plan, based on the results of laboratory tests of exhaust gases from the engine X17DTL, Opel Astra make, when operating on diesel fuel and gas fuel.

The experiments conducted make it possible to extend a database on the course of working processes in promising designs of the diesel engines, converted to gas fuel. The result of performed laboratory tests of exhaust gases from the diesel engines, re-equipped for propane-butane, is the established regularities for a change in the emissions of nitrogen oxides from the engine X17DTL, Opel Astra make, when operating on diesel fuel and gas fuel.

The obtained results for reduced emissions of nitrogen oxides when the engine operated on gas compared to diesel fuel at a change in loading by 39.7–47.1% are explained by the lower temperature of combustion of an air-gas mixture in comparison with the temperature of combustion of a diesel-air mixture. Thus, the estimated temperature of combustion for the engine X17DTL on gas was 2,207 K, and on diesel fuel – 2,346 K. The emissions of nitrogen oxides in exhaust gases of the engine at a variable crankshaft rotation frequency when operating on propane-butane is 13.9–27.5% lower compared to diesel fuel, which is explained by the 30–60% lower coefficient of excess air in gas engines compared to diesel engines.

Checking the adequacy of the mathematical model for estimating the emissions of nitrogen oxides in exhaust gases of the diesel and gas engines by comparing the calculation data with the results of experiments showed that the model satisfactorily describes emissions of nitrogen oxides. Specifically, indicators for the emissions of nitrogen oxides in exhaust gases of the diesel engine and the engine, converted to gas fuel, obtained in the experiment and calculated based on the proposed mathematical models differ by 4.6%.

The data received agree well with the results obtained in [7–9]. The proposed mathematical model could be used to assess environmental impact of exhaust gases of diesel and gas-diesel engines, though it is applicable for petrol engines. Further research will be related to the emissions of carbon oxides in exhaust gases from the diesel engines re-equipped for gas fuel.

### 8. Conclusions

1. The result of the conducted theoretical research is the construction of a mathematical model of the emissions of nitrogen oxides NO\(_x\) in the exhaust gases of engines. It is based on the basic principles of heat theory and represents the relationship between the concentration of nitrogen oxides \(r_{NO_x}\) in the exhaust gases of engines and the temperature of combustion of fuel-air mixture \(T\). A distinctive feature of this model is that it makes it possible to perform calculations of nitrogen oxides NO\(_x\) in exhaust gases of diesel engines depending on a change in the angle of rotation \(\theta\) of the engine crankshaft. The fact that the model accounts for the rate constants of chemical reactions in the combustion of gas fuel \(K_{f,\theta}\), \(K_{b,\theta}\), enables determining the emissions of nitrogen oxides NO\(_x\) in exhaust gases of the diesel engines, converted to gas.

2. Based on the performed experimental research, we have established exponential dependences of emissions of nitrogen oxides NO\(_x\) in exhaust gases of the engine on
a change in loading. The dependences were established when operating on diesel fuel and when operating on a propane-butane mixture at a fixed crankshaft rotation frequency. Given the results obtained, we can argue that the conversion of diesel engine on gas fuel reduces emissions of nitrogen oxides in exhaust gases, depending on loading, by 40–50 % on average.

3. We have established exponential dependences of emissions of nitrogen oxides in exhaust gases of the engine when operating on diesel fuel and when operating on gas at a change in the crankshaft rotation frequency. Compared to the operation on diesel fuel, the diesel engine, re-equipped for gas, reduces the emissions of nitrogen oxides in exhaust gases, depending on the crankshaft rotation frequency, by 14–28 % on average. The results obtained make it possible to optimize design of power systems for the internal combustion engines and reduce emissions of harmful substances in exhaust gases of the engines.

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