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Reduction of Emissions of Nitrogen Oxides from Traffic

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***Abstract*—**The value of emission factor was calculated in the older type of Diesel engine operating on an engine testing bench and then compared with the parameters monitored under similar conditions when the Envirox^{TM} additive was applied. It has been found out that the additive based on CeO_2 nanoparticles reduces emission of NO_x. The dependencies of NO_x emissions on reduced torque, engine power and revolutions have been observed as well.

***Keywords*—**Additive, air, cerium dioxide, emission factor, emissions, nanoparticles, nitrogen oxides

I. Introduction

AIR pollution significantly changes the natural characteristics of earth´s atmosphere and represents a serious problem. It reduces the value of assets, negatively affects the functions of ecosystems and imminently endangers human health. Contaminated air causes an increased incidence of deaths, diseases of mainly respiratory organs, reproduction and neurological disorders, cancers and diseases of circulatory system [1].

Apart from the quantitatively dominant pollutants, which include mainly CO_2, CO, SO_2, CH_4 and particulate matter (PM), air is polluted by other highly eco-toxic pollutants. The other pollutants include N_2O, O_3, heavy metals, mainly Pb, Cd, Ni and Cr, platinum metals, mainly Pt, Pd and Ru, 1,3-butadiene, benzene, toluene, o-, m-, p-xylenes, phenols, aldehydes, ketones, tar, persistent organic substances type polycyclic aromatic hydrocarbons, polychlorinated biphenyles and terphenyles, dibenzofuranes, dibenzo-p-dioxines, etc. [2]. The biggest sources of air pollution are heating plants, thermal power plants and recently mainly traffic [3].

The requirement to reduce emissions from traffic is raised by the use of cars at the expense of public transport and permanently increasing ratio of road haulage transport to rail transport [4], [5].

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The paper is focused on evaluating the quantities of NO_x emissions while applying conventional Diesel oil both without and with the additives on the basis of cerium dioxide nanoparticles (CeO_2).

II. The Analysis of Current State

The group of nitrogen oxides include nitrogen oxide NO, a colourless and odourless gas, and reddish brown nitrogen dioxide NO_2 of acrid smell, which are summarily referred to as NO_x. Other nitrogen oxides occur in the atmosphere in smaller amounts and do not represent significant risk. They include dinitrogen trioxide N_2O_3, dinitrogen tetroxide N_2O_4, dinitrogen oxide N_2O, and dinitrogen pentoxide N_2O_5. Density of the two most significant nitrogen oxides is comparable with air [6].

NO_x emissions are a serious problem, because they are linked to the burning of noble fuels (gas, Diesel oil), including biomass and their production is of a progressive nature. Cars are the primary source of up to 55 % of anthropogenic NO_x, despite catalytic converters. There are high temperatures during fuel combustion in cars. Therefore the atmospheric nitrogen N_2 oxidises to so called high-temperature NO_x [7].

NO_x reacts with ammonia, moisture, and other compounds to form nitric acid vapour and related particles. Small particles can penetrate deeply into sensitive lung tissue and damage it, causing premature death in extreme cases. Inhalation of such particles may cause or worsen respiratory diseases such as emphysema, bronchitis it may also aggravate existing heart disease [8], [9].

NO_x reacts with volatile organic compounds in the presence of sunlight to form ozone. Ozone can cause adverse effects such as damage to lung tissue and reduction in lung function mostly in susceptible populations (children, elderly, and asthmatics). Ozone can be transported by wind currents and cause health impacts far from the original sources [10].

The amount of emitted NO_x is dependent on many factors. If driving style, terrain and current weather conditions are not considered, then the NO_x emissions depend mainly on the following factors:

1) The type of engine and its technical parameters [11].
2) Principles of oxidation catalyst and its action [12].
3) The type and amount of biodiesel added to fuel [13].
4) The composition and quality of Diesel oil [11], [14].
5) The type and composition of additives added to fuel [15].

The calculation of the NO_x emissions is based on the knowledge of an emission factor Ef_m^{NOx} [g kg^{-1}] for

NO_x, which is given by formula (1) as the weight m_{NOx} [g] of the produced NO_x in relation to the weight of consumed fuel m_F [kg] [2], [16]:

$$Ef_m^{NO_x} = m_{NO_x} \times m_F^{-1} = y_{NO_x}^d \times \frac{M_{NO_x} \times n^d}{M_F \times N_F} \quad (1)$$

where M_{Nox} is the molar molecular weight NO_x [g mol^{-1}], M_F [kg.mol^{-1}] molar molecular weight of fuel, n^d [mol] substance amount of dry exhaust gases, N_F [mol] substance amount of consumed fuel, and y^d_{NOx} molar fraction of NO_x in dry exhaust gases.

There are number of additives that can be added to Diesel oil. These can be classified into three basic categories according to the character of their effects [17]:

a) Refinery additives.
b) Safety increasing and legally required additives.
c) Additives for the improvement of technical parameters and increase of fuel performance.

Refinery additives are added to fuels during production and refinery mainly in order to improve:

α) Flow properties of middle fraction and ensure that fuel does not freeze in winter season.
β) Lubricating capabilities of fuel. Additives are used to compensate for the loss of these capabilities in the production of fuels with low and ultra-low content of sulphur.
γ) Cetane number with the aim to meet the requirements for flash-point without cost demanding refinery procedures.

The safety increasing and legally required additives may be added in any phase of delivery cycle. Common additives used for these purposes are:

α) Dyes applied for red colouration of fuels in agriculture.
β) Markers for easy identification of fuels; markers do not colour the fuel, but can be extracted and then identified with the help of easy laboratory and road tests.

The additives improving the technical parameters and increasing the engine performance are added into Diesel oil in doses, usually at a distribution terminal. They may also be applied later by an end user either into a storage tank or a car fuel tank. The common additives of this category include the following:

α) Anti-foam agents reducing foam formation.
β) Additives increasing the cetane number and improving the fuel ignition properties.
γ) Anti-corrosion additives applied in storage tanks and car fuel tanks.
δ) Detergents applied with the aim to maintain fuel injection nozzles clean.

The Envirox™ Diesel oil additive should improve not only the technical parameters of fuel, but also bring a number of other advantages in comparison with conventional additives [17].

The Envirox™ is a dispersion of CeO_2 nanoparticles in aliphatic and cyclo-aliphatic hydrocarbons. Unlike conventional additives, Envirox™ is advertising efficiency throughout all combustion process, because most of the common ingredients decompose under the thermodynamic conditions prevailing in the engine combustion chamber. The additive enables engine to gain more energy from fuel and thus reduce its consumption, remove the residual soot deposits in engine combustion chamber while minimizing the formation of some contaminants [18].

Chemistry of combustion process is presented in (2) for the more efficient use of fuel energy, in (3) for the elimination of carbon deposits in engine combustion chamber and (3)-(6) for the reduction of pollutants in emissions [2].

$$(4x+2)CeO_2 + C_xH_y = (2x+y)Ce_2O_3 + xCO_2 + yH_2O \quad (2)$$

$$4CeO_2 + C = 2Ce_2O_3 + CO_2 \quad (3)$$

$$CeO_2 + Co = Ce_2O_3 + CO_2 \quad (4)$$

$$2Ce_2O_3 + 2NO = 4CeO_2 + N_2 \quad (5)$$

$$4Ce_2O_3 + 2NO_2 = 8CeO_2 + N_2 \quad (6)$$

The regeneration of CeO_2 catalyst is carried out in accordance with chemical (7):

$$4Ce_2O_3 + O_2 = 2CeO_2 \quad (7)$$

Statistically validated operational tests carried out by Oxonica company provide evidence that the recommended dosage of 5 to 10 ppm w/w CeO_2 can achieve relevant reductions in fuel consumption (by 5-12%) while reducing the emissions of CO_2, CO, NO_x, C_xH_y and particulate matter. The additive is also compatible with all standard Diesel additives [18], [19].

III. Problem Solution

A. Applied Methods and Devices

Tests to determine emission levels were carried out on the VOP-026 Sternberk company engine testing bench with the Schenk 0-900 kW electric eddy current brake operating in the range of 0-6000 revolutions min^{-1}. NM-54 Diesel was used as the primary fuel, which was mixed with 2.5×10^{-4} volumes of additives in an alternative version of comparative tests. The concentrations of CeO_2 found in Diesel oil by inductively coupled plasma atomic emission spectroscopy was 7.6 ppm w/w, which corresponds to Envirox™ suppliers' requirement.

Diesel engine with the following characteristics was used for the test: Tatra T3 930-31 four stroke, direct injection naturally-aspirated engine, air-cooled, engine cylinder capacity of 1.9×10^4 cm^3, cylinder diameter/stroke 120/140 mm, OHV distribution and a compression ratio of 1:16. The engine had 12 cylinders in two separate lines at 90°. Rated engine output was 235 kW ± 10% at 2.2×10^3 min^{-1} revolutions with a maximum torque of 1.13×10^3 N m at $1.4\times10^3 \pm 200$ min^{-1} revolutions.

The measurement of NO and NO_2 emissions was performed by a combined device called ECOM–JN for analysing the combustion gas composition, equipped with electrochemical sensor, which enabled the determination of the concentrations of CO, and O_2, which are necessary for calculating the NO_x emission factor.

Sample of combustion gas was taken by a tube probe of underpressure analyser pump. The current air mass was led from the tube probe by unheated tube to filters and moisture separator of analyser and then to pollutant sensors. It was possible to determine the concentration of CO and NO_x by applying the unheated tube between the probe and the analyser as possible combustion gas condensation on the way did not affect their concentration.

The C_xH_y content, which is also necessary for calculating the NO_x emission factor, was tested by analyser operating on the principle of flame ionization detection (FID). The principle is based on the effect that the C-H bond gets ionized during the burning of hydrocarbons in the hydrogen flame of the analyser burner combustion chamber. If the electrodes placed in the burner are energized, the value of flowing current is proportional to the number of free ions including organic matter content in the sample. The sample of gases was transported into the FID unit through a heated tube of underpressure analyser pump.

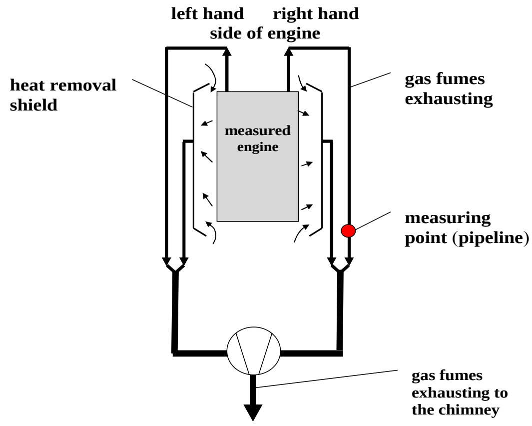

Fig. 1 Location of sampling probes

The location of measuring point, where the combustion gas velocity was measured with the Prandtl probe simultaneously with gas temperature measured with a thermocouple, is evident from Fig. 1.

B. Outcomes and Discussion

1. Mass balance

The calculation of Ef_m^{NOx} [g kg^{-1}] emission factor was based on (8) characterizing the fuel combustion, (9) for materials balance of carbon, (10) for hydrogen, (11) for oxygen, (12) for nitrogen, the amount of dry gases substance entering N_V^d [mol] and exiting n^d [mol] the combustion process, and on the measured concentration of contaminants and O_2. Fuel combustion in the engine was considered under simplified conditions in the absence of trace amounts of polycyclic aromatic hydrocarbons, N_2O, NH_3, SO_2, etc.

$$e/2N_2 + C_aH_bO_c + [a\times(1-z/2) + b/4 - c/2 + d\times(1-w/2)]O_2 = a\times(1-z)CO_2 + a\times zCO + (b/2)H_2O + d\times(1-w)NO_2 + e\times wNO \quad (8)$$

$$a\times(N_F - n_F) = n_{CO_2} + n_{CO} \quad (9)$$

$$b\times(N_F - n_F) = 2n_{H_2O} \quad (10)$$

$$2(N_{O_2} - n_{O_2}) + c\times(N_F - n_F) = n_{H_2O} + 2n_{CO_2} + n_{CO} + 2n_{NO_2} + n_{NO} \quad (11)$$

$$2(N_{N_2} - n_{N_2}) = n_{NO_2} + n_{NO} \quad (12)$$

$$N_V^d = N_{N_2} + N_{O_2} + N_{CO_2} \quad (13)$$

$$n^d = n_{N_2} + n_{O_2} + N_{CO_2} + n_{CO_2} + n_{CO} + n_F + n_{NO_2} + n_{NO} \quad (14)$$

N [mol] with corresponding subscript index in (9) – (14) represents the amount of N_2, O_2 and CO_2 entering the process, and n [mol] the amount of exhaust gases, i.e. N_2, O_2, CO_2, CO, NO_2, NO, and fuel F, exiting the process. Superscript index d means dry gases.

Substitution (15) has been introduced for easier record of other relations with symbols *a, b, c,* representing stoichiometric coefficients in (8).

$$\omega = \frac{b-2c}{4a} = \frac{\beta}{4} - \frac{\gamma}{2} \quad (15)$$

Molar fractions Y_i^d and y_i^d of i-th element in dry inlet air and exhaust gas are defined by relations (17) and (18) respectively:

$$Y_i^d = N_i \times (N_V^d)^{-1} \quad (16)$$

$$y_i^d = n_i \times (n^d)^{-1} \quad (17)$$

where N_i [mol] represents the substance amount of i-th component in the inlet air, n_i [mol] the substance amount of i-th component in output, and symbols N_V^d [mol] and n^d [mol] have the same meanings as in (13) and (14).

Equations (18) and (19) necessary for the calculation of emission factor Ef_m^{NOx} may be derived from the basis of mass balance while accepting relations (15) – (17).

$$\frac{n^d}{N_F} = \frac{a\times\left[1+\omega\times\left(1-Y_{O_2}^d\right)\right]}{Y_{O_2}^d - y_{O_2}^d + \left(1-Y_{O_2}^d\right)\times\frac{y_{CO}^d}{2} - \left(1-\frac{Y_{O_2}^d}{2}\right)\times y_{NO_2}^d - \frac{y_{NO}^d}{2} + \left[a\times(1+\omega)-(1+a\,\omega)\times Y_{O_2}^d\right]\times y_F^d} \quad (18)$$

$$n^d = N_V^d \times \frac{1+\omega\times\left(1-Y_{O_2}^d\right)}{1+\omega\times\left(1-y_{O_2}^d\right) - \frac{y_{CO}^d}{2} + (1-\omega)\times\frac{y_{NO_2}^d}{2} - \omega\times\frac{y_{NO}^d}{2} - (1+\omega)\times y_F^d} \quad (19)$$

2. Molar Fractions of Unmonitored Components

As the concentration of water vapour and CO_2 in exhaust gases were not monitored it was necessary to express their concentration from the mass balance. After modification the relation (20) has been obtained for the molar fraction in dry combustion gases y^d_{H2O} and the relation (21) for the molar fraction y^d_{CO2}, which were then used for calculating the emission factor Ef_m^{Nox}.

$$y_{H_2O}^d = \frac{\beta}{2}\times\frac{Y_{O_2}^d - y_{O_2}^d + \left(1-Y_{O_2}^d\right)\times\frac{y_{CO}^d}{2} - \left(1-\frac{Y_{O_2}^d}{2}\right)\times y_{NO_2}^d - \frac{y_{NO}^d}{2} - Y_{O_2}^d\times y_F^d}{1+\omega\times\left(1-Y_{O_2}^d\right)} \quad (20)$$

$$y_{CO_2}^d = \frac{Y_{O_2}^d - y_{O_2}^d - \left[\omega\times\left(1-Y_{O_2}^d\right)+\frac{1+Y_{O_2}^d}{2}\right]\times y_{CO}^d - \left(1-\frac{Y_{O_2}^d}{2}\right)\times y_{NO_2}^d - \frac{y_{NO}^d}{2} - Y_{O_2}^d\times y_F^d}{1+\omega\times\left(1-Y_{O_2}^d\right)} \quad (21)$$

3. Calculation of Fuel Composition

The stoichiometric coefficients in (8) can be set if the relative proportion ψ_i of each *i*-th fuel component is known. It is clear that the relations (22) and (23) are valid:

$$\beta = \frac{\psi_H\times A_C}{\psi_C\times A_H} \quad (22)$$

$$\gamma = \frac{\psi_O\times A_C}{\psi_C\times A_O} \quad (23)$$

And the relation (24) is valid for the molecular weight of fuel μ_F related to one carbon atom:

$$\mu_F = A_C + \beta\times A_H + \gamma\times A_O = 100\times A_C\times\psi_C^{-1} \quad (24)$$

where ψ_C, ψ_H and ψ_O represent the relative contents of carbon, hydrogen and oxygen in fuel; A_C, A_H and A_O [g mol^{-1}] corresponding molar atomic weights; and constants β and γ have the same meaning as in (15).

The relation (25) for constant γ is derived using (22) and (23) and (26) for constant ω after substituting γ into relation (15):

$$\gamma = \frac{\psi_O\times\left(A_C+\beta\times A_H\right)}{\left(100-\psi_O\right)\times A_O} \quad (25)$$

$$\omega = \frac{\beta}{4} - \frac{\psi_O\times\left(A_C+\beta\times A_H\right)}{2\times\left(100-\psi_O\right)\times A_O} \quad (26)$$

After substituting from relations (22) and (23) into formula (24) it applies (27) for μ_F, from which it is possible to express easily the stoichiometric coefficients *a*, *b*, *c* of (8).

$$\mu_F = \frac{M_F}{a} = \frac{100\times\left(A_C+\beta\times A_H\right)}{100-\psi_O} \quad (27)$$

4. Calculation of Emission Factor

It is possible to calculate the emission factor for NO_x Ef_m^{NOx} according to relation (28), which was deduced by substituting $n^d\times(N_F)^{-1}$ from formula (18) in (1) ad by applying the relation (27) for $\mu_F = M_F\times a^{-1}$.

$$Ef_m^{NO_x} = \frac{y^d_{NO_x} \times \frac{M_{NO_x}}{\mu_F} \times \left[1+\omega\times\left(1-Y^d_{O_2}\right)\right]}{Y^d_{O_2} - y^d_{O_2} + \left(1-Y^d_{O_2}\right)\times\frac{y^d_{CO}}{2} - \left(1-\frac{Y^d_{O_2}}{2}\right)\times y^d_{NO_2} - \frac{y^d_{NO}}{2} + \left[a\times(1+\omega)-(1+a\,\omega)\times Y^d_{O_2}\right]\times y^d_F} \qquad (28)$$

The calculated values of emission factors for NO_x under the changing conditions of engine operation when NM-54 Diesel oil was used both with and without additives are presented in Table 1. It is apparent that the NO_x emissions have been reduced by approx. 7,5% in total when additives were applied.

The total reduction of CO_2 emissions in case the additive was dosed into Diesel oil was 0,3%, which was insignificant and corresponded to the fuel consumption slightly decreased by approx. 1,5% and recorded during the whole process of measuring. Therefore it may be stated that the balance of carbon in combustion gases ranges within the uncertainty of measurement. It has also been found out that saving in fuel consumption depends on the engine operational conditions and amounts to 3% at maximum, if the reduced torque $TM_R \in\langle 890;\ 1030\rangle$ N m, engine power $P \in\langle 186;\ 214\rangle$ kW and revolutions $r \in\langle 1800,\ 2200\rangle$ min^{-1}.

The Oxonica Company has carried out long-term tests in urban and suburban transport and it advertises that the Envirox™ additive reduces NO_x emissions by up to 11% depending on the operational conditions and the type of engine, which is roughly in compliance with our findings. The reduction of CO_2 emissions should be in compliance with the reduction of fuel consumption, which was declared by the firm to be in the range of 5-12% [19]. However, this has not been proved.

The fact that the declared reduction in fuel consumption and thus the reduction in CO_2 emissions have not been proved may partially be explained by shorter time of sampling after the additive was applied in the fuel. It may also have been caused by an old type of engine produced in 1986, in case of which the above mentioned effect need not be fully displayed. Therefore further testing is planned for a newer type of engine with the sampling period after 200 hours of engine operation at minimum with the fuel containing the tested additive.

TABLE I
ENGINE PARAMETERS, CONCENTRATION OF POLLUTANTS AND NO_X EMISSION FACTOR WITH AND WITHOUT ENVIROX™ ADDITIVE

Test		ML	r	TM_R	T_{CG}	T_{EP}	P	FC	O_2	CO	NO_x	C_xH_y	$Ef_m^{NO_x}$
Fuel	No	[%]	[min^{-1}]	[N m]	[K]	[K]	[kW]	[kg kWh^{-1}]	[%]	[ppm]	[ppm]	[ppm]	[g kg^{-1}]
NM-54	1	100.0	2200	891.0	793	853	205.3	0.244	7.4	1147.2	1402.3	19.1	49.80
	2	100.0	1399	1100.0	769	828	161.2	0.213	8.1	1159.7	1410.5	16.5	31.86
	3	60.1	1803	993.0	773	833	187.5	0.226	8.4	1275.0	2083.0	18.0	48.41
	4	54.4	1799	821.6	663	720	154.8	0.220	11.0	965.0	1932.5	17.0	56.63
	5	50.0	1799	616.1	593	629	116.1	0.231	13.3	760.0	1495.0	18.0	56.83
	6	46.8	1800	408.4	527	550	77.0	0.264	15.3	602.0	1004.0	19.0	51.43
	7	100.0	2199	932.3	764	821	214.7	0.235	8.6	1262.0	1535.0	22.0	36.18
NM-54 + Envirox™	1	100.0	2201	927.8	80	840	213.8	0.236	7.7	2391.4	1905.4	36.4	41.81
	2	100.0	1399	1109.2	771	836	162.5	0.214	7.2	1770.7	1633.9	26.8	34.60
	3	63.4	1799	1022.3	767	826	192.6	0.219	8.0	1262.2	1912.7	15.3	43.21
	4	57.2	1799	817.7	673	721	154.0	0.219	11.0	846.6	1675.1	9.0	49.09
	5	53.3	1801	616.3	595	633	116.2	0.227	13.2	658.7	1279.6	8.8	47.81
	6	50.3	1801	408.0	520	549	76.9	0.260	15.3	531.7	863.9	10.7	44.35
	7	100.0	2201	934.6	772	832	215.4	0.233	7.9	2165.1	2049.9	25.1	45.81

ML [%] motor load, r [min^{-1}] engine revolutions, TM_R [N m] reduced torque, T_{CG} [K] temperature of combustion products, T_{EP} temperature of exhaust pipe, P [kW] engine power, FC [kg kWh^{-1}] fuel consumption and Ef^m_{NOx} [g kg^{-1}] emission factor for the NO_x.

At the same time the dependencies of emission factor for NO_x on the reduced torque TM_R, engine power P and revolutions r have been monitored. The example in Fig. 2 illustrates the graphical dependence of emission factors for NO_x as a function of TM_R. In accordance with theoretical expectations it may be stated that after an initial slight increase the emissions of NO_x then decrease with the increasing TM_R due to more efficient fuel combustion. Format of the trend line was evaluated by linear regression through the polynomial of second degree with the corresponding regression and reliability value R.

The increase in NO_x emissions at high TM_R while using the additive as opposed to the emissions of clear Diesel oil is difficult to be explained.

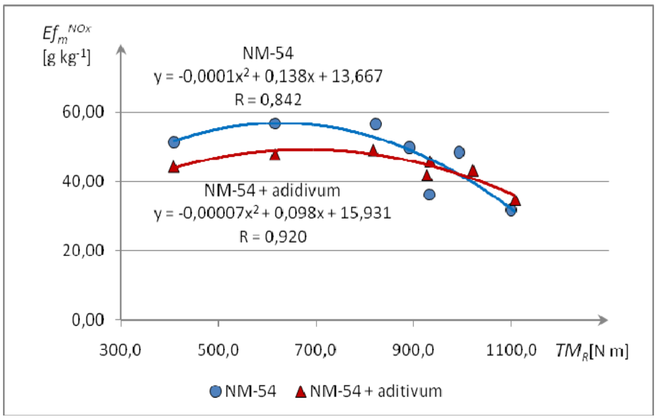

Fig. 2 Dependence of emission factor Ef_m^{NOx} on the reduced torque TM_R for both test options

Similarly the decrease in NO_x emissions was monitored at the increased engine performance. On the contrary the emission factor NO_x is the highest at engine revolutions $r \approx 1900$ min^{-1}, which is in contradiction to theory. Therefore it will be reasonable to verify the findings in further experiments under the conditions mentioned above and with a newer type of engine.

IV. Conclusion

The methodology of measuring and calculation of NO_x emissions in engine exhaust gases has been developed. It has been discovered that the Envirox™ additive based on dispersed CeO_2 nanoparticles reduces the value of the NO_x emission by approx. 7,5%, which is in compliance with the findings of the Oxonica company [19]. The above mentioned methodology may also be applied for determining the emission factors of C_xH_y, CO, CO_2 [2] and particulate matter [20]. However, the declared decrease in fuel consumption by 5-12% has not been verified. The maximum fuel savings of approx. 3% was monitored only under optimum conditions of engine operation, and was accompanied by the reduction in CO_2 emissions by approx. 2,5%.

At the same time dependencies of the emission factor for NO_x were monitored on reduced torque, engine power and engine revolutions. Beside one exception the mentioned functions were in accordance with theoretical expectations.

Disagreements with the data reported by Oxonica on fuel consumption as well as the dependencies of the emission factor NO_x on the selected engine characteristics will have to be verified in further tests on a newer type of engine and after sufficiently long period of engine operation with the Envirox™ additive.

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