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# The Influence of Metalloorganic Fuel Additives on CI Engine Emission

**Z. Chłopek[1]*, A. Darkowski[2], L. Piaseczny[3]**

[1]Warsaw University of Technology, Department of Automobiles and Building Machines,
84 Narbutta Str., 02–791 Warsaw, Poland
[2]Warsaw University of Technology, Department of Chemistry, 3 Noakowskiego Str., 00–664 Warsaw, Poland
[3]Naval University, 69 Smidowicza Str., 81–103 Gdynia, Poland

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## Abstract

The emission of particulates besides the emission of nitrogen oxides is one of the most serious problems of ecological CI engines. One means of particulates abatement is by applying catalytic additives to fuel. It suppresses particulate formation intensity and promotes the oxidation of particulates in filters. The results of influence of fuel additives (with organic magnesium) on pollution emission were presented. The studies were carried out on a Sulzer 6AL20/24 CI engine. The assessment of influence of catalytic additives to fuel on pollution was done on the basis of mathematical model analysis of the engine. It was identified that elementary emission of particulates in range of additive content lower than 0.05% is a decreasing function, and maximum abatement of emission in the considered range of engine speed and torque is about 70%, which was recognized as a very promising result.

**Keywords**: pollution emission, compression engines, fuel catalytic additives

## Important Notation

AV – average value
$b_e$ – elementary fuel consumption
**C** – vector of both exhaust concentration and elementary fuel consumption
$C_{CO}$ –volume concentration of carbon monoxide in exhaust gas
$C_{HC}$ – volume concentration of hydrocarbons in exhaust gas
$C_{NOx}$ – volume concentration of nitrogen oxides in exhaust gas
D – standard deviation
e – elementary emission
E – emission intensity
$e_w$ – relative elementary emission
g – content of fuel additives
k – light absorption coefficient
$M_e$ – torque
n – engine speed
$N_e$ – output power of engine
$r_{PM}$ – mass concentration of particulates in exhaust gas
**W** – matrix of model parameters

## Introduction

Besides nitrogen oxides, particulate emissions are one of the most serious environmental problems related to modern self–ignition or diesel engines. As announced, regulations for the admissible levels for particulates become stricter and all possible methods for their reduction must be explored. The effectiveness of catalytic fuel additives is usually characterized by their influence on soot

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*Corresponding author; e-mail zchlopek@simr.pw.edu.pl

oxidation speed and ignition temperature [1-4], although the use of ignition temperature may be somewhat misleading. In fact, the notion of ignition temperature is ambiguous in characterizing the effects on particulate oxidation and can only be used as an auxiliary quantity [1-6].

The basic requirements for all catalytic fuel additives are as follows [1]:

1. The catalyst should assure effective decrease of particulates amount in cylinders and exhaust emissions as well as increase of the oxidation intensity in the engine and in the particulates filter.
2. It is necessary to maintain typical operational properties of engines while using the catalytic fuel additives (i. e. running temperature, compression ration, detonation velocity of the air-fuel mixture).
3. If catalytic additives are not injected separately, but are added as a mixture with fuel, their chemical stability in the mixture must be kept in all conditions (i. e. stability over long time and temperatures, in small amounts of water, and with the addition of non–additive fuels).
4. Use of catalytic additives should not decrease the working effectiveness of particulate filters, i. e. covering filtering surface by metals in the additives and, by which, diminishing the field size of the active surface and increasing the exhaust gas flow resistance.
5. The use of catalytic additives cannot increase the emissions of environmentally harmful substances. Therefore, metals in the additives as well as the arising substances produced in the oxidation should not introduce a new source of nitrogen oxides and sulfur oxide emissions.

Studies of the combustion engines provide many hypothetical mechanisms for describing the effects of catalytic fuel additives [1, 2, 4, 7-16]. Often, organic and non–organic metal compounds with changing valence are used to decrease the intensity of particulates created in the cylinders and exhaust system, as well as to increase the intensity of oxidation throughout the engine's arrangements [1, 3, 4, 15-17].

The works concerning emission limitation of particulates by catalytic additives can be categorized as follows:

1. Influence of catalytic additives in lowering soot ignition temperature [1-4, 6].
2. Mechanisms of catalytic additives influence on the formation of particulates in cylinders and exhaust systems [2-5].
3. Mechanisms of catalytic additives influence on particulates oxidation in filters, research of contact soot – catalyst influence on activity of catalytic reactor [2–4, 6, 8, 13].
4. Methods of simultaneously limiting particulates and nitrogen oxide emissions [15].

Based on analysis of the present state of knowledge the following conclusions can be drawn on the topics connecting particulate emission limitation by catalytic methods:

1. The uses of catalytic oxidizing reactors makes it possible to decrease particulates emission about 30%, and simultaneously reduce both carbon monoxide emission about (40 to 90%) and hydrocarbons about (30% to 80%) [1, 5, 8, 11].
2. The application of particulate filters is a very effective method in restricting emissions of particulates. A particulate filter achieves 95% effectiveness for particles insolvable in dichloromethane (fraction INSOL [19]) and less than 30% for particles solvable in dichloromethane (fraction SOL) [6, 11]. Essential problems connected to applying this method are filter regeneration and cleaning of accumulated pollutions.
3. The addition of catalysis to fuel in such a form that it settles with particulates in filter, provides solid contact alongside soot and oxygen, ensuring catalytic reaction start when reagents reach suitable temperature [1, 3-5]. In such cases, the considerable increase of the rate of burning particulates in comparison to similar catalyst on the surface of a filter can be observed.
4. The addition of cerium to fuel causes a decrease in particulates emissions (20 to 40%) as well as a decrease in the temperature of soot ignition. This provides a favorable process for particulate filter regeneration [2, 3, 11, 13, 16, 19].
5. The application of perovskite-type oxides for catalytic soot burning causes lower soot ignition temperature, from 600°C to 320°C, and provides a favorable process for particulate filter regeneration [8, 15].
6. There is a possibility of simultaneously limiting particulate emissions and nitrogen oxides by using perovskite-type oxides [15] in addition to a metal catalyst from the platinum family [3, 12, 14].

Metal-organic compounds of iron, copper and magnesium were used to study their catalytic effects on the CI engine's emissions. The results of the organomagnesium additive are presented in this paper.

## Experimental

The effects of the catalytic additive on emission pollution control were studied on the basis of the engine's mathematical model in view of exhaust emissions, as well as fuel consumption [20, 21]. With the current state of knowledge, it was assumed [22] that concentration of exhaust components and elementary fuel consumption, in static working conditions, is functionally dependent on the engine's working conditions, characterized by:

– engine speed – n,
– torque – $M_e$.

Moreover, it was assumed that the concentration of exhaust components as well as the elementary fuel consumption were functionally dependent on the content of additive in the fuel – g.

Mathematical modeling of the engine, in view of pollution emission as well as fuel consumption, was assumed in the form of the system of equations:

$$\mathbf{C} = \mathbf{f}(n, M_e, g) \tag{1}$$

The elements of the five–dimensional vector **C** consist of both exhaust concentration and elementary fuel consumption components:

$$\mathbf{C} = \left[C_{CO}, C_{HC}, C_{NOx}, r_{PM}, b_e\right]^T \tag{2}$$

where:

$C_{CO}$ – volume concentration of carbon monoxide in the exhaust gas,

$C_{HC}$ – volume concentration of hydrocarbons in the exhaust gas,

$C_{NOx}$ – volume concentration of nitrogen oxides in the exhaust gas,

$r_{PM}$ – mass concentration of particulates in exhaust gas,

$b_e$ – elementary fuel consumption.

For testing of the mathematical model (1) experimental research results on a Sulzer 6AL20/24 engine were compared to the model (1), in accordance with Box–Behnken [23 – 25]. The structure of the model was assumed in the form of a multinomial function. A statistical analysis approach [23, 25] for the solution of the model (1) yielded a five–dimensional vectorial multinomial function with different degrees. As the final form of the model a second order multinomial was used:

$$\begin{aligned} \mathbf{C} &= \mathbf{A} + \mathbf{AM}\cdot M_e + \mathbf{AMM}\cdot M_e^{\,2} + \mathbf{AN}\cdot n + \mathbf{ANN}\cdot n^2 + \mathbf{AG}\cdot g + \\ &+ \mathbf{AGG}\cdot g^2 + \mathbf{AMN}\cdot M_e\cdot n + \mathbf{AMG}\cdot M_e\cdot g + \mathbf{ANG}\cdot n\cdot g \end{aligned} \tag{3}$$

Vectors of multinomial coefficients (3) 10x5 – dimensional matrix of model parameters – W

$$\mathbf{W} = \left[\mathbf{A}, \mathbf{AM}, \mathbf{AMM}, \mathbf{AN}, \mathbf{ANN}, \mathbf{AG}, \mathbf{AGG}, \mathbf{AMN}, \mathbf{AMG}, \mathbf{ANG}\right] \tag{4}$$

Therefore, equation (3) can be written as a five–dimensional vectorial function

$$\mathbf{C} = \mathbf{F}(n, M_e, g, \mathbf{W}) \tag{5}$$

where **W** is the matrix of model parameters of equation (3).

The range of independent variables used for modeling was established by the average condition of engine usability tests (for engine speed and torque) [24] as well as results of preliminary studies for different additive content in fuel (0% – 0.1%).

The consecutive measurement systems according to Box–Behnken, working conditions of the Sulzer 6AL20/24, as well as exhaust gas concentrations measurements are presented in Table 1. Results were determined as arithmetical averages of measurements in static engine conditions with a frequency of 1 Hz.

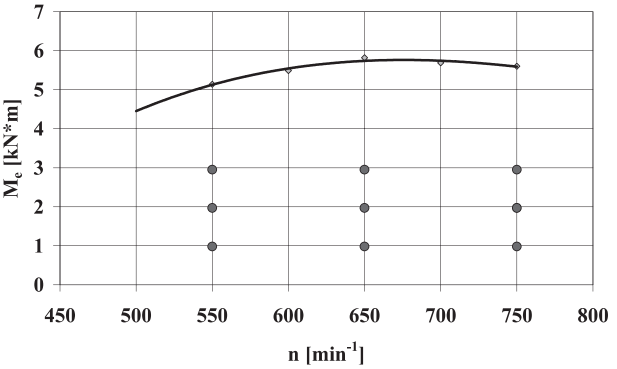

Fig. 1. External engine speed characteristics of the Sulzer 6AL20/24 engine as well as points of measurement in accordance with Box–Behnken.

Concentration measurements of the voluminal gasses CO, HC, and $NO_x$ reduction to $NO_2$ were executed by dry exhaust with analyzer working on base of the non-dispersive absorption of infrared radiation (NDIR), flame–ionic detector (FID) with heated road gas, and chemiluminescence's detector (CLD) with heated road gas, respectfully.

Mass concentration of particulates was found using analytical methods based on the measurement of absorption coefficients, executed by optical smoke meter, in accordance with equation (6) [21]

$$r_{PM} = 0.007\cdot k^2 + 0.179\cdot k - 0.004\ [\mathrm{mg/m^3}] \tag{6}$$

where: k [$m^{-1}$] – light radiation absorption coefficient.

## Results and Discussion

Table 1 presents working condition of the Sulzer engine.

For determination of the model parameters [4–6] an approximation using the Box–Benhken function (3) in four–dimensional space with three independent variables: n, $M_e$ and g was applied. The approximation was applied using a predetermined function [26], in the form of a mean square determination of previous results coupled with an error approximation function. The approximation quality is determined by the minimum of the function, in view of model parameters.

As a result, the matrix parameter **W** was determined as:

$$\mathbf{W} = \begin{bmatrix} 3.885E+02 & 2.132E+02 & -4.378E+03 & 5.396E+01 & 2.652E+02 \\ 1.229E+02 & 1.252E+01 & 1.110E+03 & 5.350E+01 & -1.287E+02 \\ 7.627E+00 & 3.645E\text{-}01 & -1.939E+02 & -3.381E+00 & 2.543E+01 \\ -3.637E\text{-}01 & -5.634E\text{-}01 & 1.510E+01 & -2.537E\text{-}01 & 5.956E\text{-}01 \\ 2.917E\text{-}04 & 4.333E\text{-}04 & -1.223E\text{-}02 & 2.553E\text{-}04 & -4.042E\text{-}04 \\ -4.495E+02 & -1.680E+02 & 3.639E+03 & -8.906E+02 & -4.672E+02 \\ 2.267E+03 & 1.133E+03 & -4.203E+04 & 1.022E+04 & -4.017E+03 \\ -2.486E\text{-}01 & -1.520E\text{-}02 & -2.905E\text{-}01 & -4.465E\text{-}02 & -4.838E\text{-}02 \\ 1.069E+02 & -1.546E\text{-}01 & -1.869E+02 & -2.727E+01 & 5.658E+01 \\ -5.000E\text{-}02 & -5.000E\text{-}02 & 1.150E+00 & -8.679E\text{-}15 & 1.150E+00 \end{bmatrix} \tag{7}$$

For the sake of postulating a physical model that was not in contradiction with the modeling object the range of independent variables could not non–zero elements. Therefore, the mathematical model of the Sulzer 6AL20/24 engine, in view of exhaust emissions and fuel consumption, has the form:

$$C_i = F_i(n, M_e, g, \mathbf{Y}_i) \geq 0 \quad \text{for} \quad C_i = F_i(n, M_e, g, \mathbf{Y}_i) \geq 0$$
$$\text{or} \tag{8}$$
$$C_i = 0 \quad \text{for} \quad C_i = F_i(n, M_e, g, \mathbf{Y}_i) < 0$$

where: i=1, K, 5;
$Y_i = \lfloor W_{ij} \rfloor$ is a row matrix for j=1, K, 5.

Figs. 2-16 show analysis results for the mathematical engine model in terms of pollution emission and fuel consumption.

The concentrations of exhaust components were determined from model (3). The intensity of individual substances emission were measured in accordance with the European Union Directive 1999/96–1/CE. To determine the model of exhaust components concentrations in an analogous way the airflow intensity in the cylinders of the engine were determined[1].

Elementary emissions of individual exhaust components (**e**) [7] was calculated by:

$$\mathbf{e} = \frac{\mathbf{E}}{N_e} \tag{9}$$

where:
E – emission intensity of individual exhaust components,
$N_e$ – output power of engine.

Using the units of measures[1] the formula (9) has the form

$$e = \frac{1}{120\pi} \cdot \frac{E}{n \cdot M_e} \ [\text{g/(kW}\cdot\text{h)}] \tag{10}$$

where:
E [g/s], n [$\text{min}^{-1}$], $M_e$ [kN·m]

Because the concentrations of exhaust components are functionally dependent on additive content in fuel, and emission intensity is a linear function of concentration, elementary emission is also a function of additive content.

$$\mathbf{e}(n, M_e, g) = \mathbf{u}(n, M_e, g) \tag{11}$$

In order to have an opinion about the influence of additive in fuel on exhaust component emissions, it is purposeful to introduce a notion of relative elementary emission in reference to elementary emission without additives to fuel

Table 1. Working conditions of Sulzer 6AL20/24 engine as well as measurement results for Box–Behnken measurements.

| Set number | $M_e$ [kN·m] | n [$\text{min}^{-1}$] | g [%] | $r_{PM}$ [$\text{mg/m}^3$] | $b_e$ [g/(kW·h)] | $C_{CO}$ [ppm] | $C_{HC}$ [ppm] | $C_{NOx}$ [ppm] |
|---|---|---|---|---|---|---|---|---|
| 1 | 0.98 | 550 | 0.05 | 0.001 | 339 | 249 | 37 | 1050 |
| 2 | 2.95 | 550 | 0.05 | 23.01 | 239 | 312 | 47 | 1395 |
| 3 | 0.98 | 750 | 0.05 | 4.97 | 361 | 199 | 34 | 889 |
| 4 | 2.95 | 750 | 0.05 | 10.36 | 242 | 164 | 38 | 1120 |
| 5 | 0.98 | 650 | 0.00 | 13.97 | 358 | 258 | 31 | 989 |
| 6 | 2.95 | 650 | 0.00 | 41.19 | 236 | 220 | 40 | 1309 |
| 7 | 0.98 | 650 | 0.10 | 26.63 | 337 | 237 | 35 | 971 |
| 8 | 2.95 | 650 | 0.10 | 48.50 | 226 | 220 | 44 | 1254 |
| 9 | 1.97 | 550 | 0.00 | 35.72 | 259 | 274 | 45 | 1358 |
| 10 | 1.97 | 750 | 0.00 | 35.72 | 248 | 183 | 40 | 1037 |
| 11 | 1.97 | 550 | 0.10 | 41.19 | 261 | 276 | 42 | 1346 |
| 12 | 1.97 | 750 | 0.10 | 41.19 | 273 | 184 | 39 | 1048 |
| 13 | 1.97 | 650 | 0.05 | 10.36 | 273 | 220 | 36 | 1424 |
| 14 | 1.97 | 650 | 0.05 | 8.56 | 276 | 219 | 34 | 1430 |
| 15 | 1.97 | 650 | 0.05 | 12.17 | 274 | 223 | 33 | 1420 |

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[1] In the paper, departures of measure units from SI standards were necessary. The reason is that these units are common in documents of European Union, as in Directive 1999/96-1/CE, (e. g. [g/(kW·h)]), and are widespread in technical environments (e. g. [$\text{min}^{-1}$]) or in Anglo-Saxon countries ([rpm]).

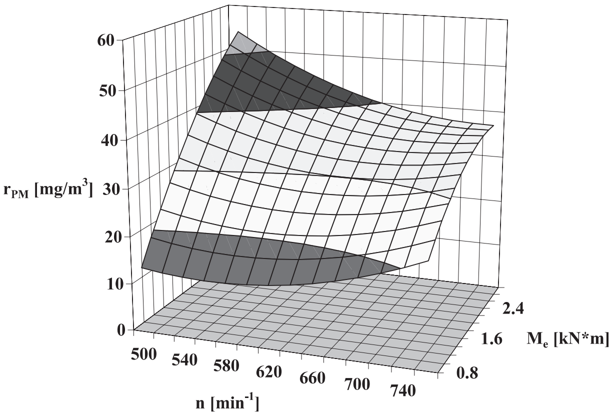

Fig. 2. Dependence of particulate mass concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0%.

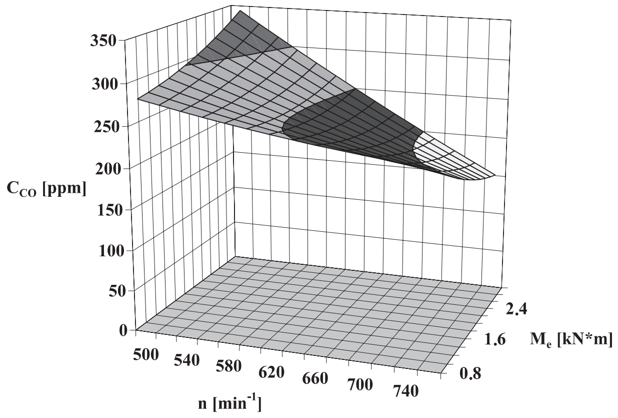

Fig. 5. Dependence of carbon monoxide concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0%.

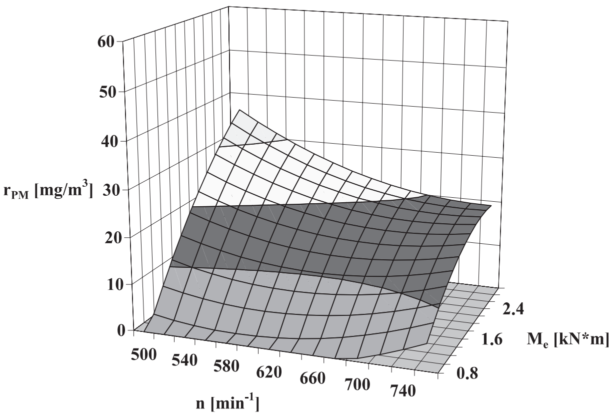

Fig. 3. Dependence of particulate mass concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0.025%.

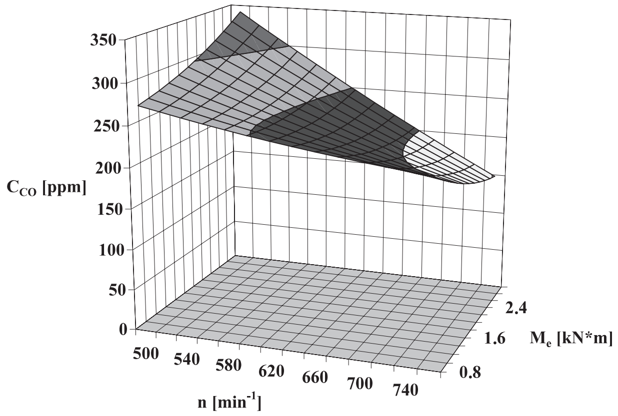

Fig. 6. Dependence of carbon monoxide concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0.025%.

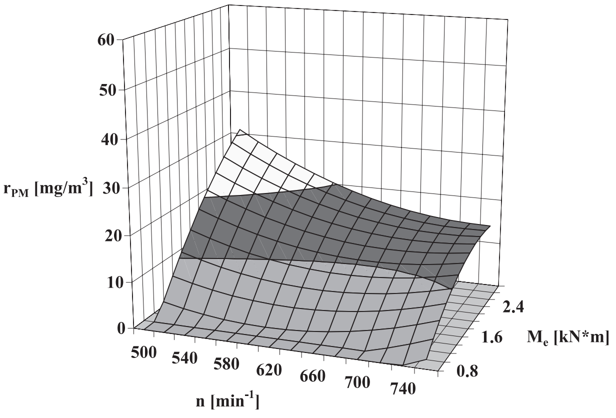

Fig. 4. Dependence of particulate mass concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0.05%.

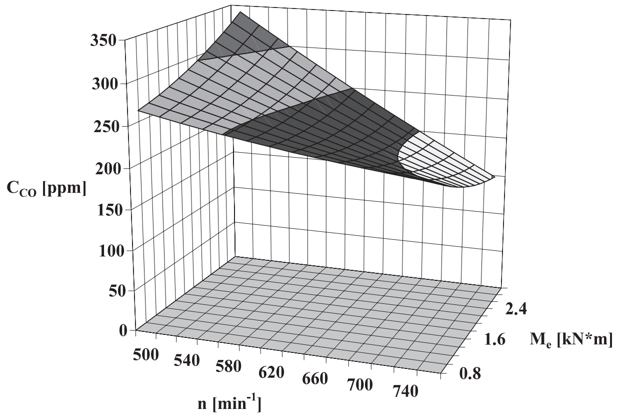

Fig. 7. Dependence of carbon monoxide concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0.05%.

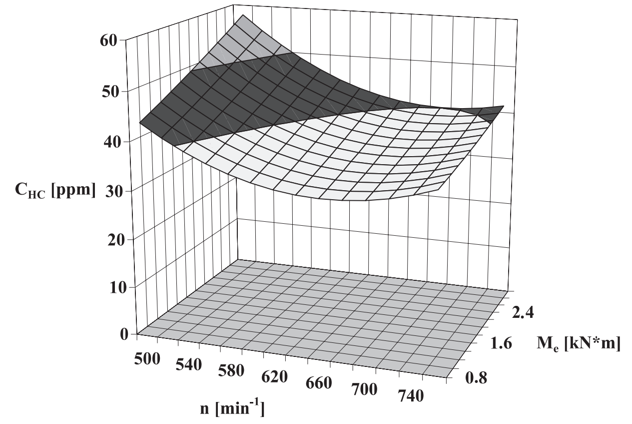

Fig. 8. Dependence of hydrocarbon concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0%.

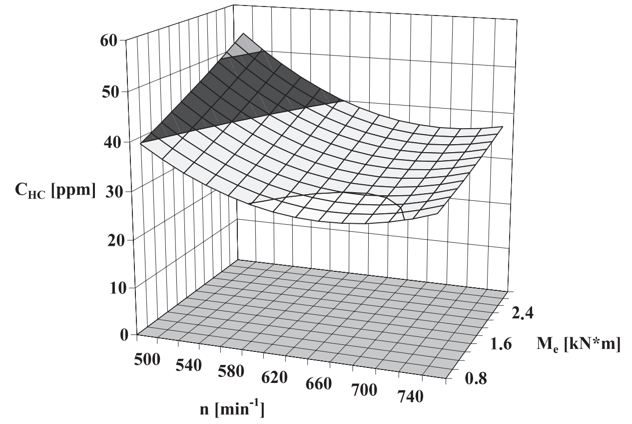

Fig. 9. Dependence of hydrocarbon concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0.025%.

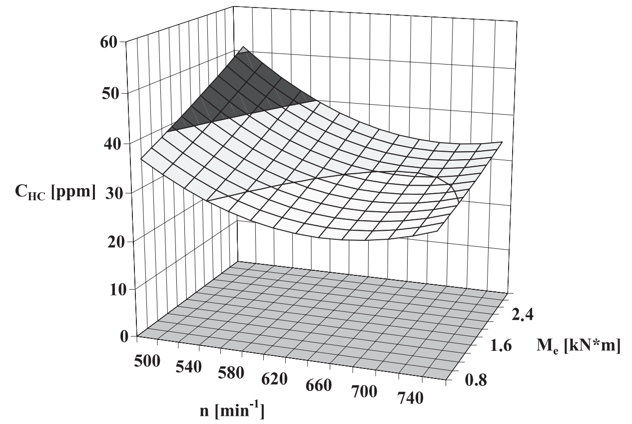

Fig. 10. Dependence of hydrocarbon concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0.05%.

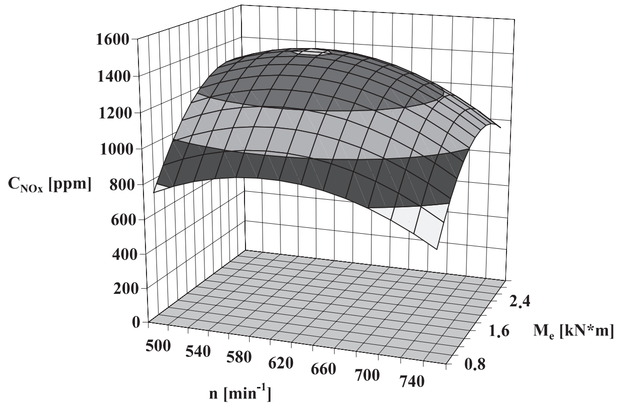

Fig. 11. Dependence of nitrogen oxide concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0%.

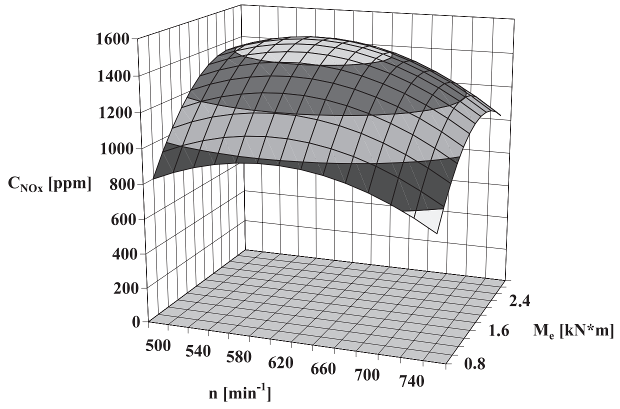

Fig. 12. Dependence of nitrogen oxide concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0.025%.

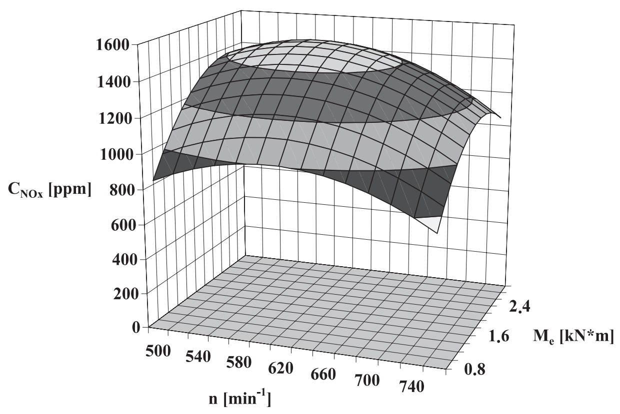

Fig. 13. Dependence of nitrogen oxide concentrations in exhaust gas on engine speed and torque for content of fuel additives g = 0.05%.

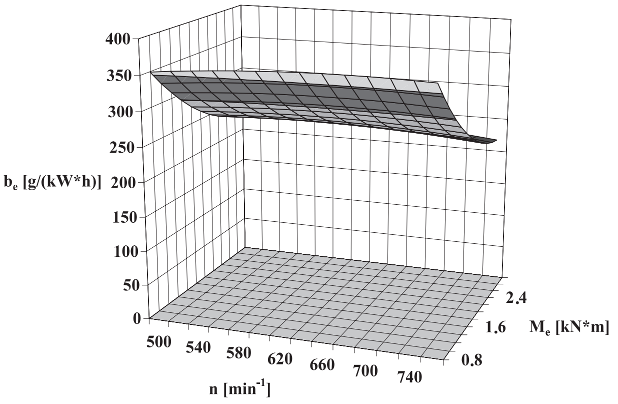

Fig. 14. Dependence of elementary fuel consumption on engine speed and torque for content of fuel additives g = 0%.

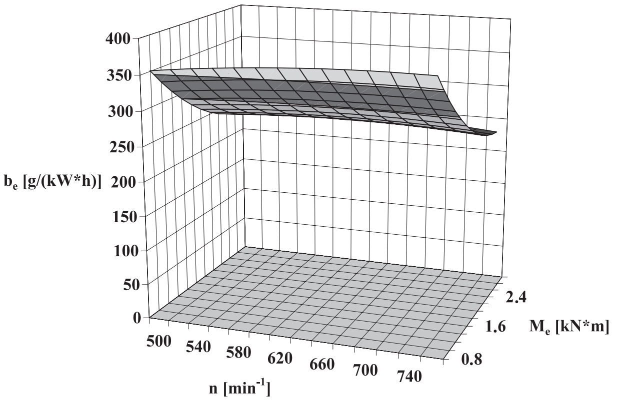

Fig. 15. Dependence of elementary fuel consumption on engine speed and torque for content of fuel additives g = 0.025%.

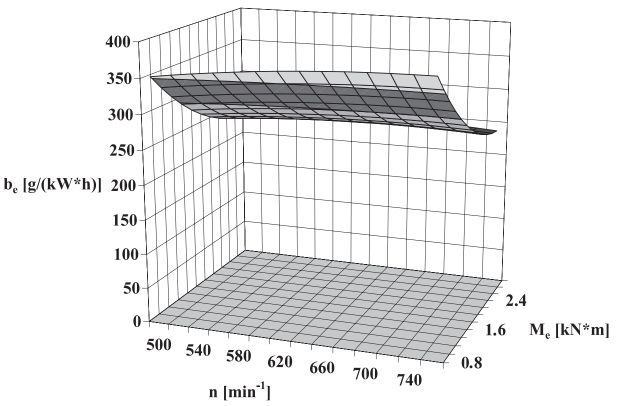

Fig. 16. Dependence of elementary fuel consumption on engine speed and torque for content of fuel additives g = 0.05%.

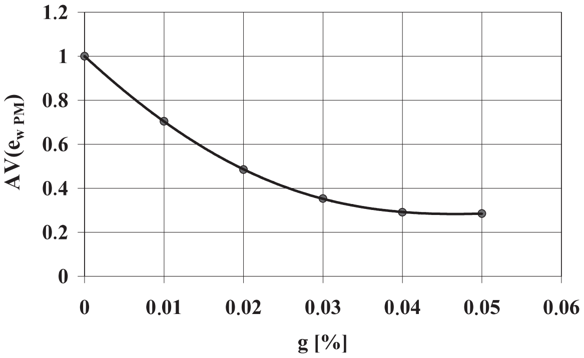

Fig. 17. Dependence of relative elementary particulate emissions on the content of studied additives in fuel.

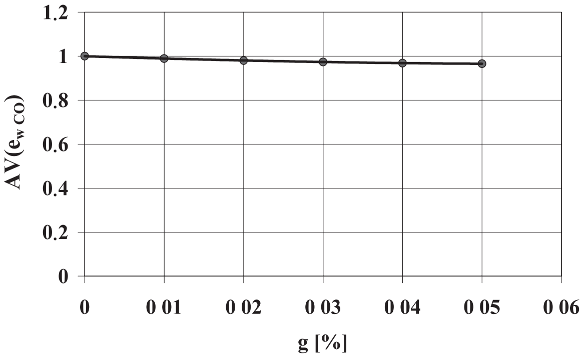

Fig. 18. Dependence of relative elementary carbon monoxide emissions on the content of studied additives in fuel.

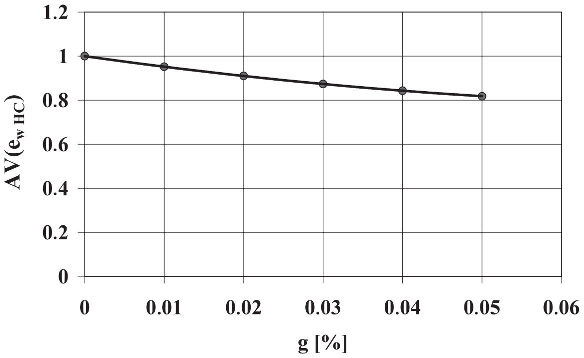

Fig. 19. Dependence of relative elementary hydrocarbon emissions on the content of studied additives in fuel.

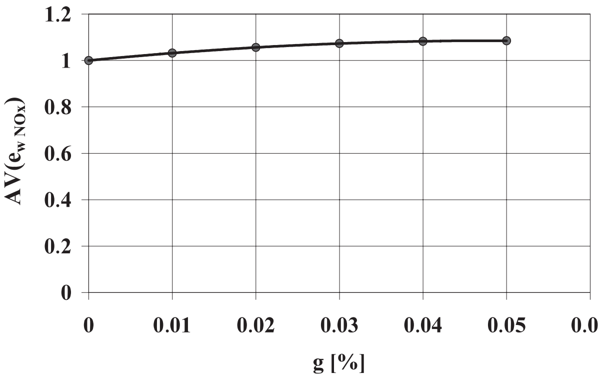

Fig. 20. Dependence of relative elementary nitric oxide emissions on content of studied additives in fuel.

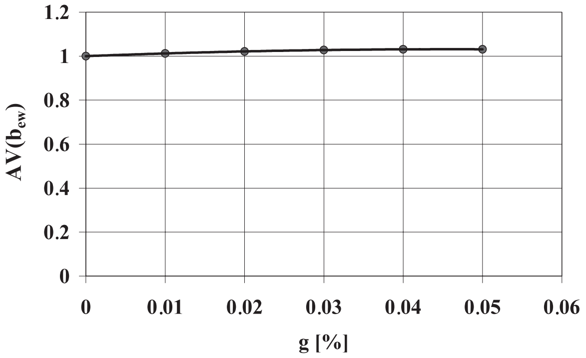

Fig. 21. Dependence of relative elementary fuel consumption on content of studied additives in fuel.

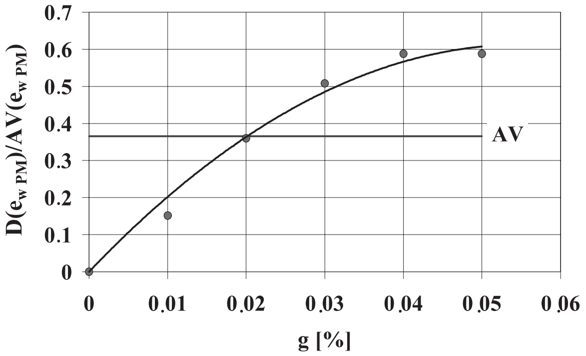

Fig. 22. Dependence of relative sensibility of elementary particulate emissions on content of studied additives in fuel.

$$e_{wj}(n,M_e,g)=\frac{e_j(n,M_e,g)}{e_j(n,M_e,0)} \tag{12}$$

where:
j = CO, HC, NOx, PM.

As a measure of influence of fuel additive on emission of exhaust components, the mean value of relative elementary emissions in the range of engine speed and torque to the limiting values in the plan of experiment was assumed. These values are:

$n_{min}$ = of 500 min$^{-1}$; $n_{max}$ = of 760 min$^{-1}$; $M_{e\,min}$ = 0,8 kN·m; $M_{e\,max}$ = 3 kN·m.

The average value of relative elementary emission in ranges of engine speed and torque reads

$$AV(\mathbf{e}_w)=\frac{1}{(n_{max}-n_{min})\cdot(M_{e\,max}-M_{e\,min})}\int_{n_{min}}^{n_{max}}\int_{M_{e\,min}}^{M_{e\,max}}\mathbf{e}_w(n,M_e,g)\,dn\,dM_e \tag{13}$$

In a similar way, the relative elementary fuel consumption and its average value in the range of considered engine speed and torque was defined.

In Figs. 17–21 dependence of relative emission of exhaust components and relative elementary fuel consumption on content in fuel of studied additive was introduced.

## Conclusions

As a result of analyses, we propose the following conclusions:

1. Particulate matter elementary emissions in the range of additive content in fuel smaller than 0.05% is a decreasing function. Maximum limitation of particulate elementary emissions in considered range of engine speed and torque is about 70%, which can be recognized as a very promising result.
2. Fuel additive in the range of content smaller than 0.05% causes almost linear decrease of hydrocarbon emissions (the maximum decrease is up to 20%).
3. Fuel additives in the range of content smaller than 0.05% cause insignificant increases of emission of remaining substances and elementary fuel consumption with tendency of insignificant decreasing of elementary carbon monoxide emission and insignificant enlarging of nitrogen oxides emission and elementary fuel consumption.
4. In the range of additive content larger than 0.05% the results of studies are less unambiguous – the growth of particulates elementary emission is observed. Formulation of conclusions in this range requires additional studies.

As a measure of relative sensibility of particulates, elementary emissions on engine speed and torque the relation of standard deviation – D and average values – AV was used – Fig. 22.

It has been confirmed that decreasing particulate emission (while increasing fuel additive content) is accompanied by higher sensibility of this effect on engine working conditions. It means that ecological results of additive application can be considerably dependent on static conditions of engine work, characteristic for its typical use.

The results of studies are very promising; particularly valuable is the possibility of considerable decrease of particulate emission by using fuel additives containing organic magnesium compounds.

In result of the analysis of engine mathematical model, and in view of the pollution emission and fuel consumption, considerable sensibility of additive influence on ecological propriety of engine has been identified. Therefore, it is necessary to analyze the conditions of engine work in relationship with real use and qualifying tests in view of emission pollution. Authors have already started studies on this subject.

Studies of other substances used as catalytic fuel additives are in progress.

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