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INCREASING THE EFFICIENCY OF INTRA-CYLINDER CATALYSIS IN DIESEL ENGINES

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The paper is devoted to the search for the rational ways of increasing the efficiency of intra-cylinder catalysis in diesel engines. The use of base metal oxides and composite oxides of transition metals as the coatings for the piston combustion chamber has been justified. This allows controlling the chemical reaction rates and reducing the formation of toxic substances. When selecting the scientific fields and rational practical ways of increasing the efficiency of intra-cylinder catalysis in diesel engines, a number of chemical, physical and technological factors were taken into consideration. The formation of catalytic coatings based on the oxides of transition metals (Mn, Co) was provided using the plasma-electrolytic oxidizing on the Al25 alloy. The coatings included both individual manganese and cobalt oxides and the mixed oxides of both metals. The catalytic coatings of the combustion chamber in the diesel engine piston on the basis of manganese and cobalt oxides and mixed oxides were tested. A maximum catalytic effect was achieved when cobalt oxides were used. The data on testing the prototype diesel showed that the fuel rate in this case was decreased by 4–6%, the emission of NO_x was reduced by 14–15% and the emissions of carbon oxide were reduced by 20–25%.

Keywords: diesel engine, oxide coating, intra-cylinder ecological catalysis, combustion chamber, piston, plasma electrolytic oxidizing, catalytic activity, toxic substances.

Introduction

The current level of ecological indices of diesel engines was achieved by the multiple investigations which were aimed to improve the operating process by increasing the efficiency of fuel supply systems and gas–turbine supercharging system, using flue gas neutralizing, purification and recirculation agents etc. A promising approach to a further improvement of the ecological indices of diesel engines is considered to be the intra-cylinder ecological catalysis. Catalytic processes result in a more perfect fuel combustion and conversion of the products of incomplete combustion (carbon monoxide CO and hydrocarbons C_nH_m) and the toxic components of solid particles (SP) into innocuous substances and also a reduction in the emission of nitrogen oxides (NO_x).

The analysis of the research performed to study the intra-cylinder ecological catalysis in diesel engines shows the conflicting character of the influence of coatings including the metals of a platinum group on NO_x emissions and the products of incomplete fuel combustion.

The paper [1] gives the research data on the influence of platinum coating applied onto the piston surface. A certain biasing of the period of fast combustion to the top dead center (TDC) was observed, the emission of C_nH_m was reduced by 5%, the CO emission was decreased by 10% and the combustion completeness was improved. However, the concentration of NO_x in flue gases was increased by 10%. The drawback of catalytic coatings made of precious metals is their high cost.

The paper [2] deals with the investigations of coatings made of copper, nickel and chromium applied on combustion chamber walls. The obtained data showed that all the coatings provided a decrease in the fuel rate and CO and C_nH_m emissions. At the same time, the emission of NO_x was increased. Copper thereat turned out to be a more efficient catalyst.

Iron, cobalt and manganese oxides and other materials including spinels, perovskites and hexaaluminates [3] can be used as the coatings. For example, the paper [4] gives the research data for

the six-cylinder diesel with the gas-turbine supercharging and catalytic MgZrO_3 coating of pistons; cylinder heads and valve heads were coated with CaZrO_3 . The fuel rate was reduced by 1–2% as a result of the selection of an optimal duration of the injection of fuel and heat-insulation material. The SP emission was reduced by 40% and the NO_x emission was reduced by 10%.

The authors of the paper [5] assessed the effect of two-layer catalytic coatings, deposited on the combustion chamber surface, 0.15 mm NiCrAl and 0.35 mm $\text{Y}_2\text{O}_3\text{-ZrO}_2$, on the operation of diesel with the gas-turbine supercharging in the range of crankshaft speed of 1100 to 2000 min^{-1} . The results of this research showed that the engine power was increased by 2% and the effective fuel consumption was reduced by 4.5–9.0%, the flue gas opacity was reduced approximately by 2%, and CO emissions were decreased by 9%, however, the emission of NO_x was increased by 10%.

Use of the oxide ceramic coatings of base metals allows improving the combustion process due to an increase in temperature and a decrease in the ignition delay time, and reducing cooling system heat losses and decreasing emissions of unburnt C_nH_m , CO and SP. In spite of an increase in the actuating medium temperature, a decrease in NO_x emissions is observed [6].

The authors of [7] worked out two options of the application of catalytic coatings onto the piston combustion chamber surface. For the first option, the nichrome layer Cr20Ni80 with a thickness of 0.15 mm was applied using the gas-plasma deposition. The second option differed by the composition: Cr15Ni80Al5 powder layer with a thickness of 0.15 mm is deposited onto the substrate. It was noted that the NO_x concentration was reduced by 25% with a simultaneous reduction in the engine operating severity; a relative decrease in CO within 10–20% was registered in the entire range of diesel operation at $n=1500 \text{ min}^{-1}$ and the soot content was reduced by 20%.

Thus, the research data obtained earlier give us an opportunity to assume that the use of base metal oxide and composite oxides of transition metals as combustion chamber coatings allows us to affect physical and chemical processes in the diesel engine cylinder and create the conditions for complete fuel combustion and control the reaction rate in order to reduce the formation of harmful substances in the cylinder.

To consider catalytic conversion of the products of incomplete fuel combustion and NO_x in the diesel engine cylinder, some chemical, physical and

technological factors should be taken into account. Chemical factors are related to an increase in the rate of chemical reactions due to an increase in the number of catalytically effective centers on the combustion chamber surface in the zones of fuel accumulation [8].

Technological factors are related to the specification of optimal conditions for the application of catalytic coatings. It was already mentioned that it is reasonable to use the oxides of transition metals (Mn, Co, Fe, Cu etc). Aluminum alloys, in particular Al25, are considered to be the most widely spread materials used for the fabrication of the pistons for diesel engines. We are interested in the coating formation modes that enable the oxidation of aluminum with the simultaneous application of catalytically active systems based on the transition metal oxides. To provide the adhesion interaction with the substrate and obtain the coatings of a substantial thickness with the developed surface, the high modes of anodizing are used [9]. The surface treatment of metals using the plasma electrolytic oxidizing (PEO) enables the formation of multifunctional coatings with a broad range of properties, in particular, mechanically strong, wear resistant, catalytically active, electroinsulating, heat and corrosion resistant ones, diesel soot combustion [10] etc. The PEO is the most effective method that can provide the formation of catalytic coatings on the combustion chamber surface that are made of valve metals (Ti, Zr, Mg, Al) and their alloys. A minimum thickness of the catalytic coating contributes to the regular removal of soot formation from the piston combustion chamber surface due to the self-regeneration of the surface of catalytic coating.

Physical factors have an impact on catalytic processes by providing the mixing quality that is characterized by fuel supply indices, air charge motion and the combustion chamber shape. Creation of the high-inertial flow of air charge enables reinforcement of the contact of fuel vapors that are burnt under the action of catalyst on the combustion chamber surface [11]. One of the defining physical factors is considered to be a shape of the combustion chamber of piston. The qualitative mixing is mainly provided by fuel supply characteristics for the diesel engines with the direct injection of fuel and the fuel injection equipment that provides the maximum pressure of injection up 150 MPa. However, to improve the catalytic effect we must strive for the enlargement of the surface of combustion chamber (coating) mainly due to the enlargement of the side wall of displacer and formation of the stepwise groves

and edges, as shown in Fig. 1.

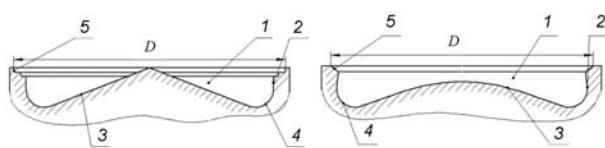


Fig.1. Shapes of the combustion chamber of diesel engines with the maximum pressure of injection up 150 MPa:

1 – the combustion chamber; 2 – the side wall;
3 – the chamber bottom; 4 – the radius of transition from the side surface to the chamber bottom; 5 – the turbulization edge

The previous research data enabled the development of methodological prerequisites for the formation of PEO coatings on titanium and aluminum alloys, including, highly doped ones [3,12]. It was shown [3] that water electrolytes based on the pyrophosphates of alkaline metals meet the most of the requirements. Their use enables the formation of metal oxide catalysts of the combustion of hydrocarbon fuel based on transition metal oxides, including, cobalt oxides that possess a high thermal stability and have oxygen affinity; these demonstrate considerable advantages at high operating temperatures [13]. At the same time, the opportunities for the synthesis of oxide coatings of combined composition were actually not realized and we expect the manifestation of synergetic effect for them those results in a considerable increase of the reactivity. The purpose of the research was to justify the practical ways of increasing the efficiency of intra-cylinder catalysis in diesel engines.

Experimental

The coatings were formed on substrates of aluminum AK12M2MgN alloys (Al25). The surface pretreatment of the samples was performed by mechanical cleaning, degreasing, etching and rinsing with distilled water. The composition of electrolytes and parameters of oxide systems synthesis are given in Table 1.

Table 1
Composition of electrolytes and the parameters of oxide systems synthesis

No.	Substrate	The composition of electrolyte		Current density i , A/dm ²
		Components	Concentration, mol/dm ³	
1	Aluminum alloy	K ₄ P ₂ O ₇	0.400	3.0–5.0
		CoSO ₄	0.100	
2	AK12M2MgN	NaOH	0.005–1.000	10.0–20.0
		KMnO ₄	0.050–0.200	

The acidity of the electrolytes was maintained

in the pH range of 9–11.5. The electrolytes were prepared from certified reagents graded as «pure grade» and «analytical grade» with the use of distilled water.

The PEO of substrates was conducted using a B5-50 stabilized power supply that maintained voltage up to 300 V. PEO was carried out in an electrolytic cell under conditions of forced cooling of the electrolyte (temperature of 25–30°C) and stirring the electrolyte.

The chemical composition of the coatings was determined by means of X-ray photoelectron spectroscopy using INCA Energy 350 energy dispersive spectrometer. X-ray radiation was excited via exposing the samples to a 15-keV electron beam. In addition, X-ray fluorescence analysis was conducted using a SPRUT portable spectrometer with a relative standard deviation of 10⁻³–10⁻²; the error in determining the component content was ±1 wt.%.

The surface morphology of the coatings was studied using Zeiss EVO 40XVP scanning electron microscope (SEM). The SEM images were recorded by the registration of secondary electrons (BSEs) via scanning with an electron beam; this mode allows studying the topography with a high resolution and contrast ratio.

The surface morphology and roughness of coatings were studied by atomic force microscopy AFM using NT-206 microscope. Scanning was performed using the contact probe CSC-37 (lateral and vertical resolutions 2 and 0.2 nm, respectively; 1024×1024 scanning matrix, CSC cantilever B as probe, probe tip radius of 10 nm).

To estimate the influence which the catalytic coating of the surface of combustion chamber in the piston has on the combustion process behavior and the catalytic conversion of toxic substances, we carried out the investigation of single-cylinder diesel 1CH12|14 in terms of load characteristic at a crankshaft rotation frequency of 1400 min⁻¹.

The experimental unit included the load device, starting electric motor, fuel-flow meter, air-flow meter of the Bosh Company, the control equipment, thermocouples and the sensors. The tests were performed using the standard diesel fuel at actually similar atmospheric conditions. The gas analyzer «OXI-5M» was used for the measurement of NO_x and CO concentrations and the smoke opacity was measured using the «Hartridge» smoke meter of a MK-3 model.

Results and discussion

Chronograms of inter-electrode voltage in the PEO formation of oxide coatings on titanium and

aluminum alloys have a classical form and are divided into the following characteristic regions: pre-spark, spark, micro-arc and arc discharges [12].

The forming voltage grows rapidly in the first 2–3 minutes of PEO process. The dependence $U-t$ is almost linear in the pre-spark region for all electrolytes and substrates due to the formation of barrier oxide Al_2O_3 .

The beginning of the sparking corresponds to the breakdown of dielectric films. The growth of the voltage is slowing down due to compete processes of breakdown and regeneration of barrier oxide. At this stage, thermochemical reactions begin that provides incorporation of electrolyte components into the forming oxide layer. The inclusion of the dopants in the oxide systems changes the surface morphology of the synthesized materials.

The PEO process stabilizes in the region of stable sparking. The incorporation of transition metal oxides into the surface layers at the stable PEO is manifested in the appearance freeform sites of different colors: violet for cobalt and black for manganese.

Peculiarities of PEO modes for Al alloys

It is established that the quantitative characteristics of the PEO stages, such as inter-electrode voltage U_f , sparking voltage U_s and maximum forming voltage U_{max} , depend on the nature of the treating substrate and the composition of the electrolyte.

The most significant inclusions of dopants in the oxide layers (Table 2) are achieved in micro-arc mode.

Table 2

The composition of oxide coatings

Mixed oxide system	The contents of components, wt.%
Co-containing	O 26–55; Al 33–49; Si 1.8–2.8; Co 8.7–23.6
Mn-containing	O 61–62; Al 2.75–3.85; Si 0.4–0.9; Mn 33–36

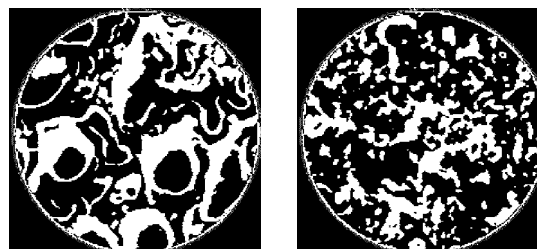
Transition metal oxides gradually and uniformly cover the surface of the treated sample (Fig. 2). The processing voltage in micro-arc stage remains almost unchanged.

It is the micro-arc mode that is optimal for the synthesis of uniform oxide coatings with the highest content of dopants (Table 1) for aluminum alloy.

The transition of the oxidation process to the arc discharge region causes the oxide layer destruction by arcs of high-intensity fields.

Ceramics-like mosaic surface structure of mixed

oxides obtained on the AK12M2MgN alloy in the electrolyte no. 1 consists of agglomerates sphere-like islet structures of non-stoichiometric cobalt oxides (Fig. 4,a). PEO treatment of AK12M2MgN alloy in the electrolyte no. 2 (Fig. 4,b) allows obtaining surface layer of micro-globular structure containing non-stoichiometric manganese oxides [14].



$AlSAI_2O_3 \cdot CoO_y$

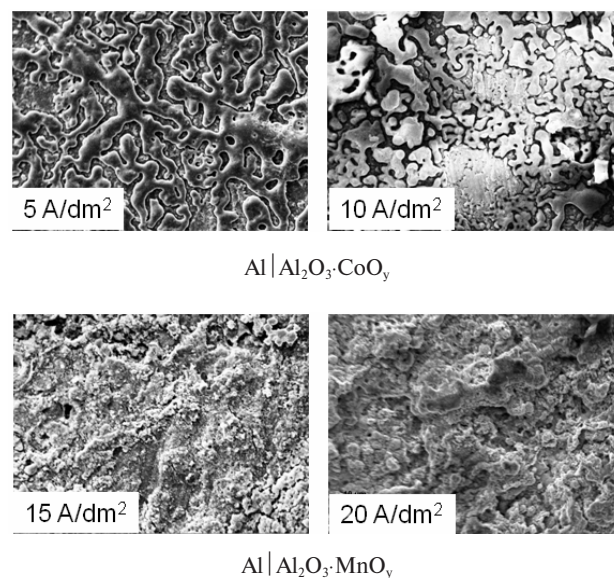
$AlSAI_2O_3 \cdot MnO_y$

Fig. 2. Morphology of oxide systems obtained from electrolytes (Table 1) during 30 min of PEO treatment.

Magnification $\times 1000$

Varying the operating current density also affects the morphology of the surface coatings (Fig. 3). This is explained by appropriate changes in the oxidation rate and duration of PEO stages.

The high degree of oxide coatings surface development during the PEO of aluminum alloys confirms by the results of surface topography analysis using scanning probe microscopy [14].



$Al | Al_2O_3 \cdot CoO_y$

$Al | Al_2O_3 \cdot MnO_y$

Fig. 3. Effect of current density on the coatings surface morphology. Magnification $\times 500$

The composition, surface morphology and surface topography of mixed oxide systems

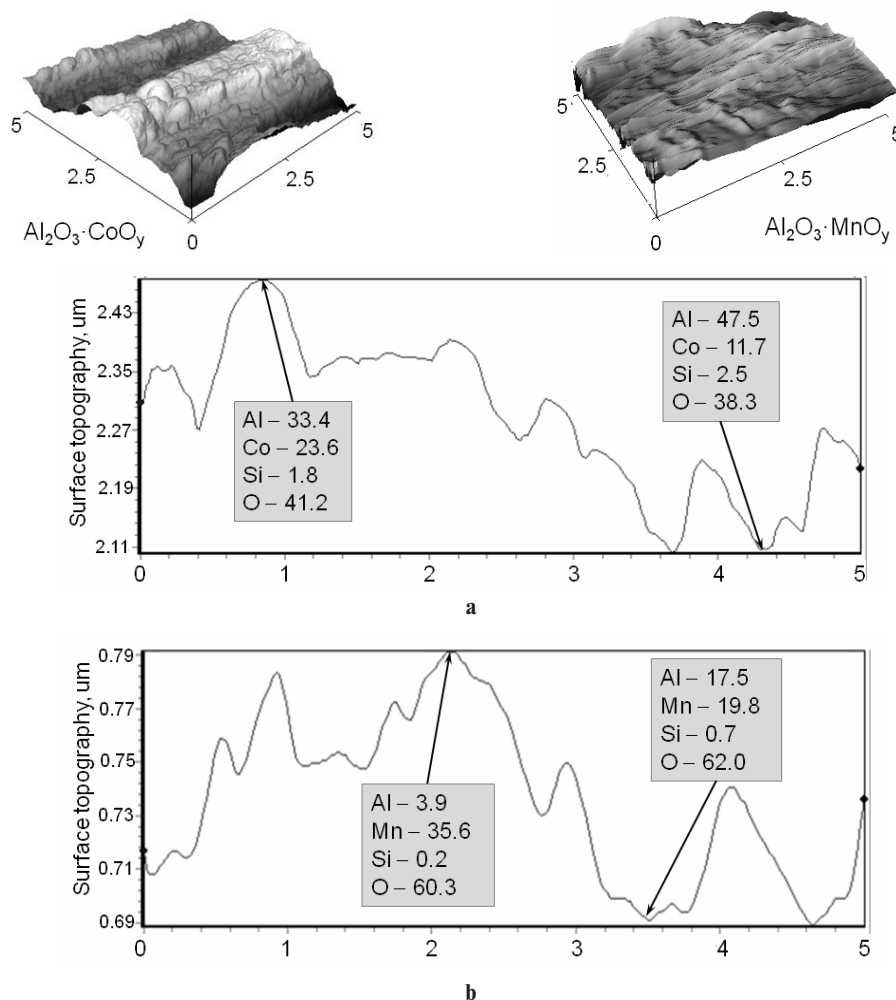


Fig. 4. 3D maps and profiles of the surface for mixed oxide systems:
a – Al|Al₂O₃·CoO_y; b – Al|Al₂O₃·MnO_y. Scanning area 5×5 mm

Al|Al₂O₃·CoO_y, Al|Al₂O₃·MnO_y predetermine high catalytic activity in red-ox reactions.

Influence of the catalytic coating of the surface of combustion chamber in the piston on diesel engine indices

The obtained data are shown in the following coordinates: hourly fuel consumption (G_f) as a function of torque (M_t) (Fig. 5) and the concentrations of nitrogen oxides (W_{NO_x}) and carbon oxide (W_{CO}) in flue gases as a function of torque (M_t) (Fig. 6).

The analysis of the relationship between hourly fuel consumption and the torque moment shows that the catalytic cobalt oxide-based coating on the surface of combustion chamber in the piston allows increasing the fuel efficiency by 4–6%.

In case of catalytic cobalt oxide-based coating, the concentration of NO_x is reduced and at maximum loads this reduction reaches 14–15% in comparison with the uncoated piston of a commercial design

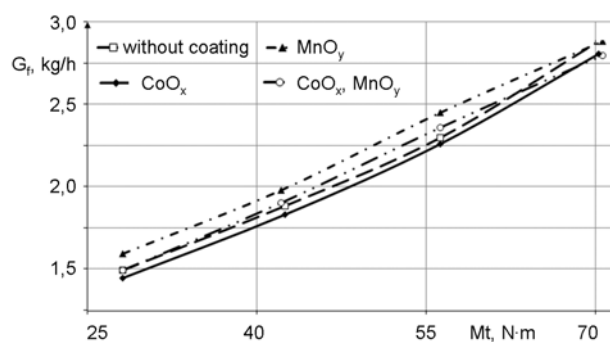


Fig. 5. Hourly fuel consumption (G_f) plotted as a function of torque (M_t) at $n=1400 \text{ min}^{-1}$

and catalytic coatings based on manganese oxide and mixed cobalt and manganese oxides.

The reduction in the CO concentration to 29% is observed in case of catalytic coatings based on manganese oxide and cobalt oxide, however, CO

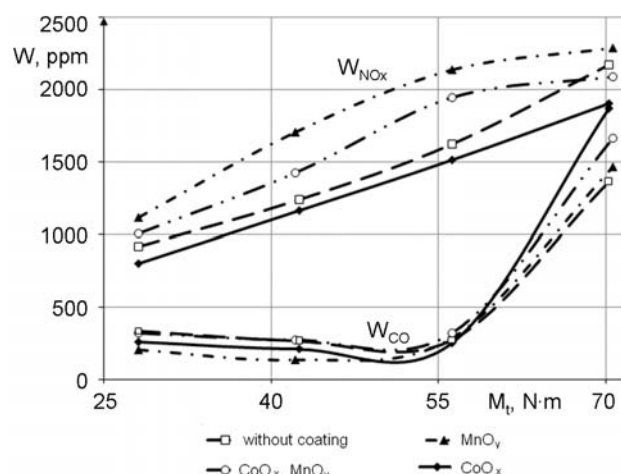


Fig. 6. Concentrations of nitrogen oxides (W_{NOx}) and carbon oxide (W_{CO}) in flue gases plotted as functions of the torque (M_t) at $n=1400 \text{ min}^{-1}$

emission diminishes only to the values of excess air ratio $\alpha=1.7$.

Conclusions

1. The research was performed in order to systematize and select scientific fields and rational practical ways for increasing the efficiency of intracylinder catalysis in the diesel engine. The obtained results showed that the application of oxide coatings for the combustion chamber using base metals and the composite oxides of transition metals allows affecting physical and chemical processes in the diesel engine cylinder, creating the conditions for complete fuel combustion and controlling the reaction rates to reduce the amount of formed harmful substances.

2. When taking into account the physical factors, it is necessary to provide the selection of the shape of combustion chamber with a maximum possible surface alongside with the appropriate fuel supply indices and the air charge motion; the reduction of heat losses in the combustion chamber wall will be compensated in this case by the effect of partial heat insulation provided by the substrate and applied catalytic coating.

3. The procedure of the formation of catalytic coatings based on transition metal oxides (Mn, Co) was suggested that implies the use of plasma – electrolytic oxidizing of Al25 alloy in the presence of oxo-anions and cations of a different nature. That may provide a further improvement of electrochemical and thermochemical reactions of intermolecular redox conversions and allows forming the layer of catalytically active material with a tight adhesion to the substrate.

4. The results of the investigation of the catalytic coatings of combustion chamber based on manganese oxides, cobalt oxides and mixed oxides carried out

for the ICH12|14 diesel allowed us to conclude that the cobalt oxide coatings on the combustion chamber of aluminum piston provides the highest catalytic effect. The application of the CoO_x coating onto the surface of combustion chamber allows improving the fuel efficiency by 4–6% and reduce the emission of nitrogen oxides by 14–15%; the emissions of carbon oxides are reduced by 20–25% reaching the values of excess air ratio $\alpha=1.7$.

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Keywords: diesel engine; catalytic coating; intra-cylinder ecological catalysis; combustion chamber; piston; harmful substances; catalytic action; toxic substances.

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