RESEARCH ON THE USE OF BIOGAS AS AN ADDITIVE TO COMPRESSED NATURAL GAS FOR SUPPLYING VEHICLE ENGINES

Oleksandr Kravchenko[1,*], Roman Symonenko[2], Juraj Gerlici[1], Andrii Golovan[3], Serhii Shymanskyi[4], Igor Gritsuk[5], Yuriy Grytsuk[6]

[1]University of Zilina, Zilina, Slovakia
[2]Center for Scientific Research of Complex Transport Problems, State Road Transport Research Institute, Kyiv, Ukraine
[3]Department of Navigation and Maritime Safety, Odesa National Maritime University, Odesa, Ukraine
[4]Sector for Ensuring the Circulation of Cards for Digital Tachographs, State Road Transport Research Institute, Kyiv, Ukraine
[5]Department of Operation of Ship Power Plants, Kherson State Maritime Academy, Odessa, Ukraine
[6]Department of General Engineering Training, Donbas National Academy of Civil Engineering and Architecture, Ivano-Frankivsk, Ukraine

*E-mail of corresponding author: avtoap@ukr.net

Oleksandr Kravchenko 0000-0003-4677-2535,
Juraj Gerlici 0000-0003-3928-0567,
Igor Gritsuk 0000-0001-7065-6820,
Serhii Shymanskyi 0000-0001-5316-2466
Roman Symonenko 0000-0002-4269-5707,
Andrii Golovan 0000-0001-6589-4381,
Yuriy Grytsuk 0000-0003-3389-1172,

Resume

Internal combustion engines, which operate in various load and speed conditions, are significant sources of environmental pollution that affect a vehicle's movement in traffic. Automotive engines typically operate in unsteady-state modes, transitioning cyclically from one operating mode to another during vehicle operation. The potential of using biogas as an additive to compressed natural gas for automotive engines was explored within this research. The results of experimental studies on the fuel efficiency and environmental performance of a vehicle with a petrol engine, converted to run on a mixture of compressed natural gas and biogas, are presented. A comparative analysis was conducted on the fuel and economic performance, as well as environmental indicators of A-92 petrol, compressed natural gas, and a mixture of compressed natural gas and biogas in the European driving cycle modes.

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1 Introduction

The escalating energy demands of humanity result in increased energy consumption, leading to resource depletion and environmental pollution. Currently, the pressing issue is to conserve natural resources and protect the environment while simultaneously increasing energy production to meet human needs. This can be achieved by utilizing alternative fuels derived from renewable sources. Biogas can be produced through the anaerobic decomposition of organic waste.

Fully or partially purified biogas is similar to compressed natural gas (CNG) in terms of its characteristics. As a result, it can be stored and transported in a compressed or liquefied state [1]. Special tankers can deliver it to petrol stations, and it can be supplied through the special gas pipelines.

Biogas is composed of methane (CH_4), carbon dioxide (CO_2), small amounts of carbon monoxide (CO), hydrogen (H_2), nitrogen (N_2), oxygen (O_2), water vapor (H_2O), and hydrogen sulphide (H_2S). When used as a fuel, biogas affects the operation of internal combustion engines differently than the traditional fuels.

According to a literature review, most studies have focused on evaluating the environmental impact, fuel efficiency, and engine performance of biogas [2] and CNG as alternative fuels compared to traditional gasoline. Khan et al. [3] stated that using biogas as an additive to CNG results in the same fuel economy and lower carbon dioxide emissions compared to vehicles fueled solely with CNG. Bordelanne et al. [4] found that vehicles fueled with biomethane-enriched CNG can reduce the greenhouse gas emissions by more than 80% compared to gasoline-powered vehicles. Agbulut et al. [5] discovered that adding biogas to CNG can reduce exhaust emissions and improve performance metrics in gasoline engines. Chandra et al. [6] observed that methane-enriched biogas showed almost similar engine performance compared to compressed natural gas in terms of brake power output, specific gas consumption, and thermal efficiency. Shamekhi et al. [7] stated that CNG, may improve the fuel efficiency and environmental performance of gasoline engines compared to traditional fuels like gasoline. Singhal et al. [8] suggested that biogas is a potential fuel for providing a continuous supply of CNG in the form of bio-CNG, which contains about 95% methane. Lemke et al. [9] found that converting a production vehicle to run on gasoline, CNG, and biomethane shows potential for improving fuel efficiency and environmental performance compared to traditional fuels like A-92 gasoline. Melaika et al. [10] observed that gaseous fuels, such as natural gas and biogas, can improve engine efficiency, reduce standard emissions and particulates, and provide additional benefits like lower particulate numbers compared to gasoline. Wargula et al. [11] observed that CNG-fueled engines reduced fuel consumption by 31% and hourly estimated machine operating costs resulting from fuel costs by 53% compared to traditional gasoline engines. Limpachoti and Theinnoi [12] found that Compressed Biomethane Gas (CBG) fuel in a spark ignition engine has higher thermal efficiency and lower NOx and HC emissions compared to CNG. Pavlovskyi [13] have shown how the use of biodiesel blends with rapeseed oil additives can improve fuel efficiency and reduce emissions in diesel engines, specifically noting a significant reduction in total mass emissions when using three-component biodiesel fuels.

However, the impact of using biogas as an additive to compressed natural gas on the fuel efficiency and environmental performance of gasoline engines has not been extensively researched, particularly when compared to traditional fuels like A-92 gasoline. The gaps in research discussed above motivate us to focus on answering the question - "What is the impact of using biogas as an additive to compressed natural gas on the fuel efficiency and environmental performance of gasoline engines converted to run on this mixture compared to traditional fuels such as A-92 gasoline?" The research question aims to investigate the impact of compressed natural gas and biogas blends on the environmental performance and fuel efficiency of vehicle engines. The study results will provide valuable insights into the potential of biogas as an alternative fuel that can help reduce harmful emissions and increase energy independence.

The paper's structure can be summarized as follows. Section 2 provides a detailed methodology for studying the impact of using biogas as an additive to compressed natural gas on the fuel efficiency and environmental performance of petrol engines. Section 3 presents and discusses the study results. Section 4 presents conclusions and prospects for further research.

2 Research methodology

A methodology for researching the impact of biogas on engine and vehicle performance has been developed for conducting experimental and theoretical studies.

The research methodology comprises theoretical and experimental measures. Conclusions can be drawn about the effectiveness of using a mixture of natural gas and biogas to improve the fuel, economic, and environmental performance of a vehicle in operation [14].

Figure 1 shows the flowchart of this methodology, which comprises eight stages.

The initial stage includes an analysis of previous studies on the use of biogas to reduce pollution from cars and save oil products. This stage requires analyzing research conducted by individual authors and research organizations both in Ukraine and abroad. When analyzing the literature, biogas was evaluated to determine its advantages and disadvantages [15]. Special attention was given to the potential for its widespread use as a motor fuel and its ability to improve the environmental performance of vehicles [16]. According to the analysis in the first section, it is predicted that in the near future, a mixed fuel consisting of CNG, and biogas will likely replace petrol as a fuel for spark-ignition automotive engines, rather than pure biogas.

The second stage entails selecting and justifying a test-driving cycle, conducting experimental research on regulating the car engine power supply system while driving a car in the European urban driving cycle, and improving gas fuel equipment [17].

At the third stage, the permissible content of the inert component (6%) was determined by simulating a mixture of CH_4 and different proportions of CO_2. Based on the characteristics and properties of the available biogas, the ratio between the components of the mixed fuel - CNG 80% and biogas 20% was determined. The similarity between the proposed mixed fuel and the simulated gas was established. Subsequently, due to the lack of an adequate amount of biogas, large-scale experimental studies of the car were carried out when the engine was powered by the simulated gas.

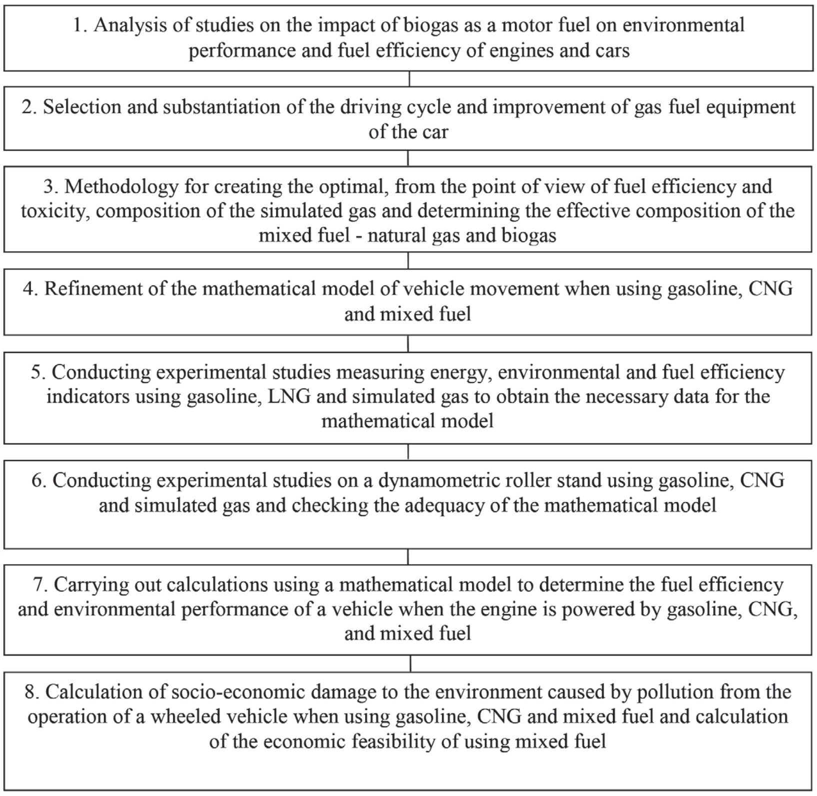


Figure 1 *Flowchart of the general research methodology*

Mathematical modelling is frequently employed to determine engine performance across a wide range of operating conditions, including transient modes [18]. Therefore, it is necessary to refine the existing mathematical model [19] that simulates the movement of a car during test cycle modes to study the efficiency of using different types of fuels, particularly mixed fuels (stage four).

When compiling a mathematical model of vehicle movement according to the European driving cycle modes, seven modes are distinguished, according to the types of their identical mathematical description [20-21]:

- engine operation in idling mode;
- engine acceleration in idling mode;
- vehicle starting from a standstill in the first gear;
- gear shifting from higher to lower and from lower to higher;
- vehicle movement at a constant speed;
- vehicle deceleration with the clutch locked;
- engine deceleration with the clutch disengaged.

Mathematical dependencies describing changes in the main parameters characterizing the engine operation in the study were compiled based on the results of experimental studies. The description of the parameters was carried out by a factorial experiment in a wide range of changes in speed and load modes on a dynamometric roller stand. As a result, hourly fuel and air consumption and concentrations of harmful substances in engine exhaust gases were established [20].

For calculations based on the refined mathematical model, it is necessary to conduct a number of experimental studies to measure the fuel efficiency and environmental performance [22] when the car engine is powered by petrol, CNG and simulated gas (similar to mixed fuel) and to develop polynomial dependencies that describe engine performance in a wide range of speed and load conditions when powered by petrol, CNG and mixed fuel (stage five).

The mathematical model was improved to calculate the fuel consumption and the concentration of harmful substances in engine exhaust gases under different speed and load conditions during the driving cycle. The model was tested using petrol, CNG, and mixed fuel (stage six).

During the seventh stage, experimental tests were conducted on the car using petrol, CNG, and simulated gas to determine its environmental performance and fuel efficiency in different driving modes over the driving cycle. The tests were carried out on a dynamometer roller stand under engine conditions. Additionally, the obtained data from the mathematical model is compared to the experimental data.

At the eighth stage, the socio-economic damage to the environment [23] caused by pollution from the operation of a wheeled vehicle using petrol, CNG and mixed fuel was calculated, and the economic feasibility [24] of using mixed fuel was calculated.

Currently, there are three groups of driving cycles used worldwide to assess the fuel efficiency and environmental performance of vehicles: European, US, and Japanese driving cycles. For research purposes, it is appropriate to use the European driving cycle of a car, as regulated by UNECE Regulation No. 83. This regulation provides uniform provisions for the approval of vehicles regarding the emission of pollutants in accordance with engine fuel requirements.

The European Driving Cycle, also known as the EU and ECE (European Economic Commission) test cycle, is a program designed to closely simulate urban driving. It was supplemented in 1993 by the Extra Urban Driving Cycle (EUDC), which includes suburban driving at speeds of up to 120 km/h. The New Urban Driving Cycle (NEDC) is a combination of the urban and suburban cycles.

The use of natural and biogas as motor fuels has some disadvantages compared to liquid fuels. These include a low energy concentration in 1 m^3 of the combustible mixture, which results in a decrease in the effective power of the engine. For the spark ignition engines, the decrease is around 11-12% with optimization of the ignition advance angle and 17-19% without optimization.

For combustible mixtures with an excess air ratio of $\alpha = 1$, lower calorific value of 1 m^3 is: petrol-air - $h_H = 3739$ kJ/m^3; gas-air - $h_H = 3404$ kJ/m^3; and biogas-air (with $CH_4 = 62\%$) - $h_H = 2168$ kJ/m^3.

Based on the preliminary calculations, the heat reduction in 1 m^3 of the engine fuel mixture is 8.7% when using natural gas, 42.1% when using biogas, and 15.25% when using a mixture of 80% natural gas and 20% biogas.

To prevent a decrease in engine energy performance and energy losses during the biogas purification when used as a motor fuel, it is recommended to use it as a mixed fuel with natural gas.

Experimental studies were conducted on a vehicle with an external mixture formation engine. (Table 1), which was converted to run on gas fuel. The car was tested using petrol, CNG, and a mixture of CNG and biogas. For petrol, a carburetor-mixer was used, while for CNG, a three-stage reducer, gas metering, and carburetor-mixer were used. The fuel-air mixture in the engine was regulated by the vacuum in the inlet pipeline. Feedback on the oxygen content in the exhaust gases of the engine was not used.

The studies remain relevant as there are still many

***Table 1** Technical characteristics of the car*

Title	Value
Fuel	A-92
Engine displacement, cm^3	1300
Engine power, kW	52
Engine torque, Nm / Engine speed, rpm	96 / 3400
Engine layout / number of cylinders	straight / 4
Maximum speed, km/h	145
Fuel consumption (combined cycle), liters per 100 km.	9
Fuel consumption (urban cycle), liters per 100 km.	11
Fuel consumption (suburban cycle), liters per 100 km.	8
Compression ratio	8.5
Engine crankshaft moment of inertia, kg m^2	0.15
Wheel moment of inertia, kg m^2	0.7
Dynamic wheel radius, m	0.255
Transmission	4 manual transmission (mechanics)
Gearbox ratios	
- 1 gear	3.753
- 2 gear	2.303
- 3 gear	1.493
- 4 gear	1
Final drive ratio	4.3
Drive type	Rear
Front suspension	Independent. Multi-link
Rear suspension	Dependent. On longitudinal levers
Front brake	Disk
Brake rear	Drum
Tire size	155 SR13
Curb weight, kg	955

vehicles in operation that run on both petrol and gas fuels without exhaust gas oxygen feedback.

The experiments were conducted on a simulated roller stand (Figure 2), strictly adhering to the predetermined speeds, to accurately measure the level of harmful substances present in the exhaust gases (CO, NO_x, CH, C_mH_n, CO_2).

Several experimental studies were conducted to determine the feasibility of using the biogas mixed with CNG as a motor fuel. The studies required a substantial amount of biogas.

Large-scale biogas production has not yet been established in Ukraine. Biogas is mainly produced in the industrial and agricultural sectors. In the private sector, biogas is produced in smaller volumes and under non-standardised conditions, resulting in fluctuations in biogas characteristics.

The enterprises use the biogas they produce, but taking constant samples or large volumes of gas would disrupt the closed production cycle. This would not be suitable for the producers as it would interrupt the biogas production process. Consequently, it was not possible to purchase enough biogas for testing.

It is possible to construct a biogas plant independently. However, due to the small size of the plant, the composition of the biogas may vary at the beginning, middle, and end of the production process. A stable biogas composition is crucial for the road transport.

The availability of biogas may change over time if a network of gas filling stations that sell biogas according to the standard is established, or if it is produced in large volumes and is consistent across filling stations from production to consumption. Convincing evidence of the efficiency of biogas production and use as a motor fuel is required for this to occur, which is the focus of this research.

Due to the current shortage of biogas that could be used for experimental studies of automobile engines, a methodology for simulating gas containing methane and carbon dioxide in certain proportions (CH_4 - 94% and CO_2 - 6%) was developed, which corresponds to a mixture of CH_4 - 80% and biogas 20% with a CO_2 content close to 6%.

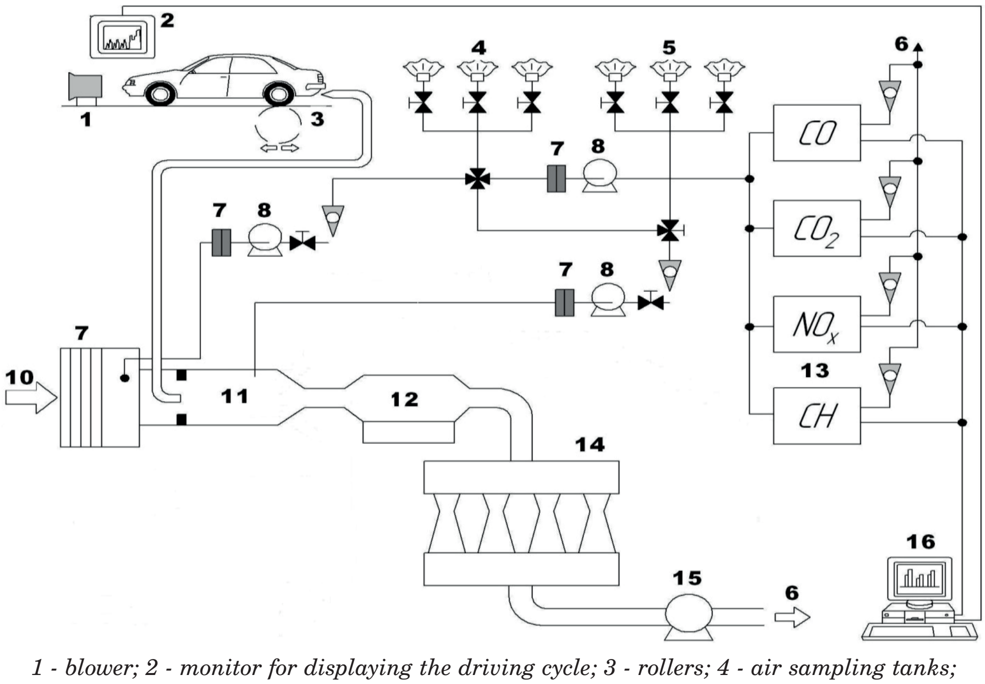
The required content of the inert component was determined by simulating a mixture of CH_4 with varying proportions of CO_2.

Two methods of adding CO_2 were used in the sequence of experiments:

1) adding CO_2 to the mixture with air, and
2) adding CO_2 to a mixture with CNG.

To determine the optimal dose of CO_2 and the appropriate method of addition, the stand was equipped with a reference volumetric gas meter called "CALIBR", with factory number 00017. The meter has a measuring range of 0.016 to 10.00 m^3/h with an error of ± 0.3% of the measured value.

The purpose of the set of experimental studies of the car is to determine the effect of using different types of fuels to power the engine: A-92 petrol, CNG and simulated gas (which is close to mixed fuel in its



1 - blower; 2 - monitor for displaying the driving cycle; 3 - rollers; 4 - air sampling tanks; 5 - exhaust gas sampling tanks; 6 - exhaust gas outlet; 7 - filter; 8 - vacuum pump; 9 - heater; 10 - air supply; 11 - exhaust gas dilution tunnel; 12 - heat exchanger/heater; 13 - gas analyzers; 14 - venturi nozzles; 15 - centrifugal vacuum pump; 16 - computer for processing results

Figure 2 *Diagram of the simulating roller stand* composition), on its performance in different load and speed modes and in the modes of the European driving cycle.

To accomplish this objective, the following tasks were completed:

1. Optimal adjustment of gas equipment, considering the improvement of the vehicle's environmental performance and the improvement of gas fuel equipment.
2. Checking the compliance of the simulated gas with the mixed fuel in the modes of the car's movement according to the European driving cycle.
3. Determination of the energy, fuel-economic and environmental performance of the engine when operating in different load and speed modes and active idling mode when powered by different types of fuels.
4. Determining the performance of the car when the engine is powered by A-92 petrol, CNG and simulated gas during the testing on a dynamometer roller stand to verify the adequacy of the refined mathematical model.

A factorial experiment was conducted to assess the economic and environmental performance of the engine using different types of fuels and advanced gas cylinder equipment.

The dynamometer continuously recorded the following parameters during the tests: vehicle speed, total fuel consumption in gallons per mile (G_{pal}), concentrations of carbon monoxide (CO), total hydrocarbons (CH), non-methane hydrocarbons (C_mH_n), nitrogen oxides (NO_x), and carbon dioxide (CO_2) in dilute exhaust gases.

The calculation method was utilised to determine the specific G_{CO} - mass emissions of carbon monoxide per cycle, G_{CO2} - carbon dioxide, G_{CH} - total hydrocarbons, G_{CmHn} - non-methane hydrocarbons, and G_{NOx} - nitrogen oxides.

During the testing, exhaust gases are continuously analysed and sampled using the Constant Volume Sampling method and stored in elastic containers. The emissions of harmful substances are calculated by averaging the concentrations of harmful substances in the exhaust gases. This is obtained by processing a dataset of instantaneous values of concentrations of harmful substances in the exhaust gases, or by averaging the concentrations of harmful substances in diluted exhaust gases sampled into an elastic container.

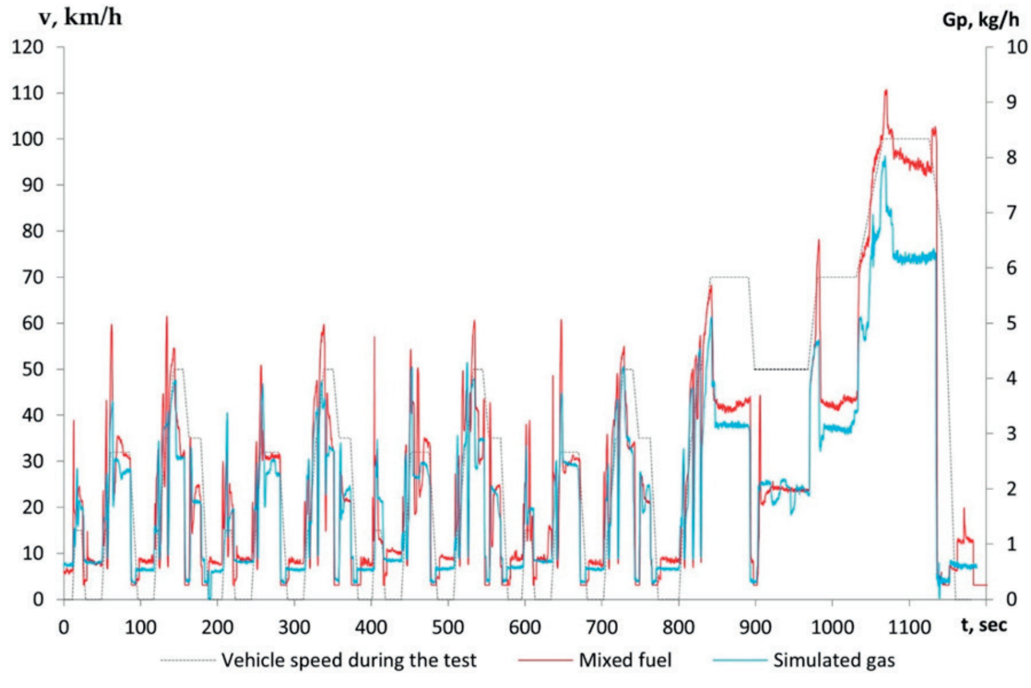


Figure 3 *Instantaneous fuel consumption values (Gp) during the car test on the roller stand in the driving modes according to the European driving cycle*

Table 2 *Fuel consumption and harmful substances emissions during the testing of a car on a dynamometer roller bench in the European driving cycle modes*

Fuels	Fuel consumption, g/cycle	Mass emissions of harmful substances			
		G_{CO}, g/km	G_{NOx}, g/km	G_{CO2}, g/km	G_{CmHn}, g/km
Mixture of natural and biogas	720.56	0.5313	0.4552	113.2	0.3463
Simulated gas	677.35	0.5040	0.4269	106.4	0.3334

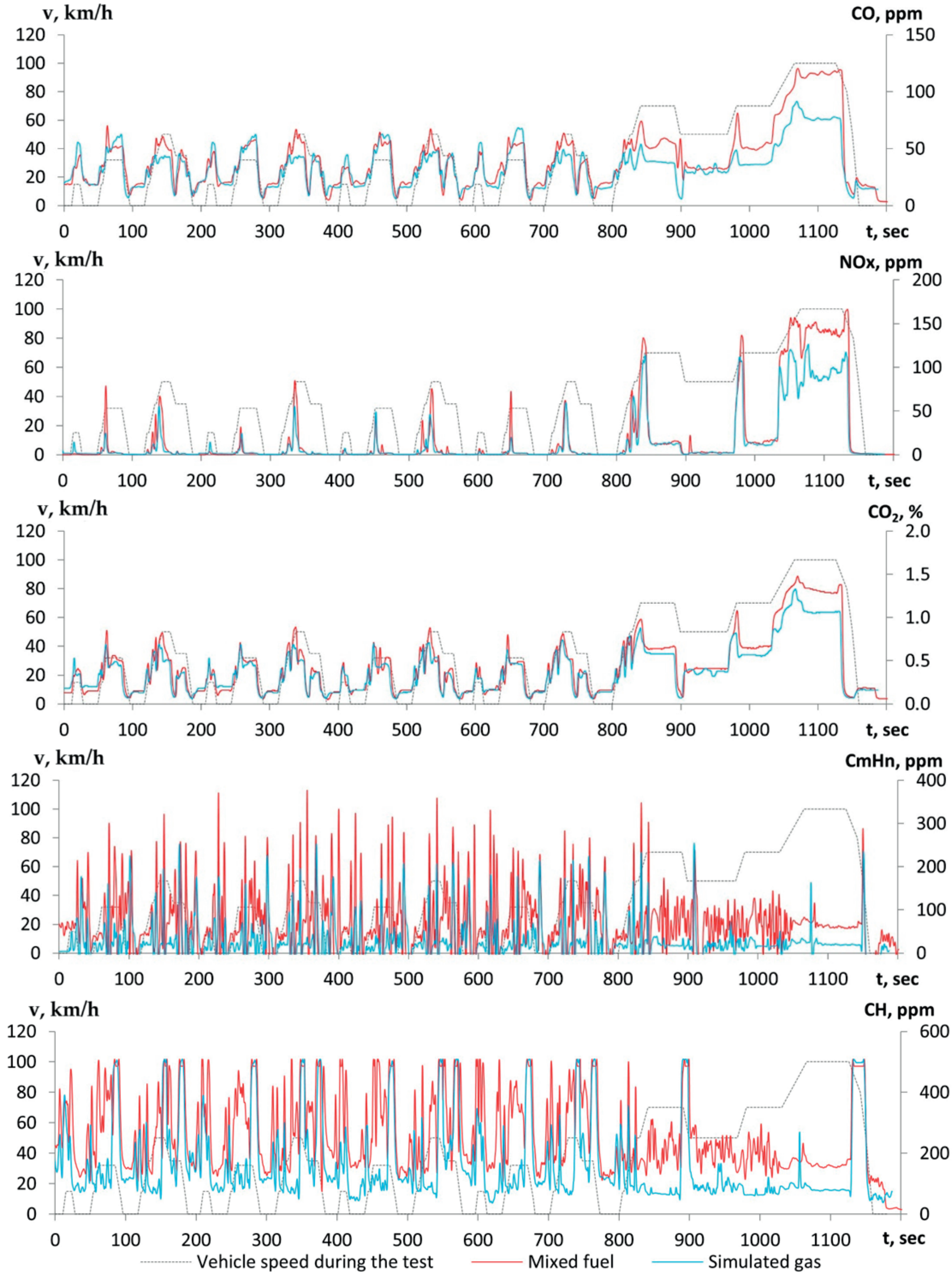


***Figure 4** Instantaneous concentrations of harmful substance oxides in exhaust gases during the test of a car on a dynamometer roller test bench in driving modes according to the European driving cycle*

3 Research results

Tests were conducted to evaluate the environmental performance of the engine using different types of fuel: petrol, CNG, mixed fuel (80% LNG and 20% biogas) and simulated gas (94% LNG and 6% CO_2).

The biogas used in the research has the following composition: CH_4 - 57%, CO_2 - 30%, N_2 - 11%, H_2O - 3%, O_2 - 0%.

The results of the first stage of testing of the vehicle are shown in Figures 3-6.

An excessive level of carbon monoxide CO concentration in exhaust gases, when the car engine is powered by CNG, indicates excessive enrichment of the gas-air mixture. The increase in CO concentration occurs in the engine braking mode.

The level of NO_x concentrations with exhaust gases, when powering an CNG engine, is almost the same as with petrol.

Emissions of total hydrocarbons CH increase in the engine braking mode. The level of concentrations of non-methane hydrocarbons C_mH_n in the exhaust gases of the engine when powered by CNG is reduced compared to petrol.

Figure 3 displays the results of measuring instantaneous fuel consumption during a car test on a roller bench in the European driving cycle modes when the engine is powered by a mixture of fuels (80% CNG and 20% biogas) and simulated gas (94% CNG and 6% CO_2). Figure 4 shows the results of measuring instantaneous concentrations of harmful substances in the exhaust.

The results of the tests of the car, when the engine is powered by mixed fuel and simulated gas, on a dynamometer roller stand in the driving modes according to the European driving cycle, are given in Table 2.

According to the results of tests of the car on a dynamometer roller stand in driving modes according to the European driving cycle, when the engine is powered by mixed fuel, fuel consumption increases by 6% compared to the engine power supply with simulated gas.

The emissions of harmful substances in exhaust gases are reduced when simulated gas is used compared to mixed fuel. Specifically, carbon monoxide (CO) is reduced by 5.1%, nitrogen oxides (NO_x) by 6.2%, carbon dioxide (CO_2) by 6%, and non-methane hydrocarbons (H_{CmHn}) by 3.7%.

The experimental studies confirm that the simulated gas is suitable for large-scale experiments using mixed fuels.

Thus, a simulated gas (CH_4 - 94% and CO_2 - 6%) was used for a large series of experimental studies, which corresponds to a mixture of CH_4 - 80% and biogas 20% with a CO_2 content close to 6%. The biogas used in the research has the composition: CH_4 - 57%, CO_2 - 30%, the rest are gases that do not participate in the combustion process (N_2 - 11% and H_2O - 3%).

Figure 5 displays the instantaneous fuel consumption values obtained during the car test on a roller stand in the European driving cycle.

Figure 6 displays the measurements of harmful substance concentrations in engine exhaust gases during car testing on a dynamometer roller in the European driving cycle.

Tables 3 and 4 present the results of tests conducted on the car using various types of fuel on a dynamometer roller stand in the European driving cycle.

Based on the results of the car's tests on

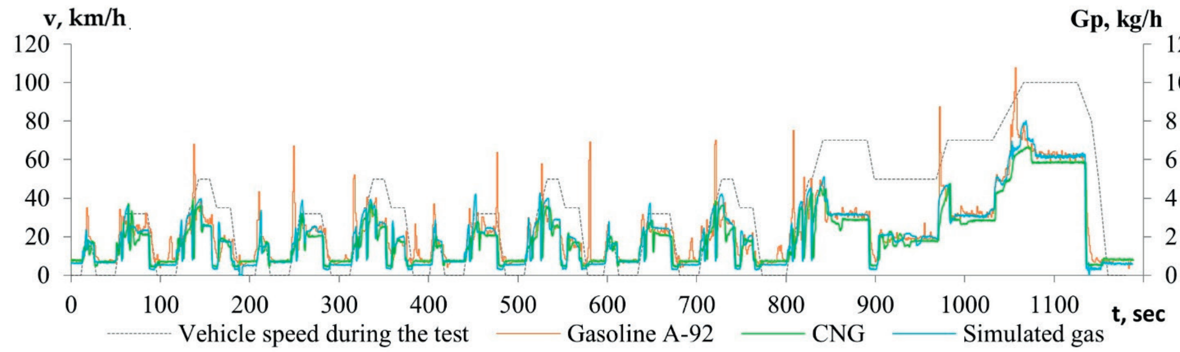


***Figure 5** Instantaneous values of fuel consumption G_{fuel} during the test of the car on the roller stand in the European driving cycle modes*

***Table 3** Fuel consumption during testing of the car on a dynamometer roller bench in driving modes according to the European driving cycle*

Fuels	g/cycle	MJ/cycle	Fuel consumption
A-92 petrol	750.81	33.04	9.17 litters/100 km
CNG	652.61	30.02	7.96 m^3/100 km
Simulated gas	677.35	29.26	7.8 m^3/100 km

a dynamometer roller under the conditions of the European driving cycle, it was found that the fuel consumption (g/cycle) decreased by 13.1% when powered by CNG and by 9.8% when powered by simulated gas, compared to A-92 petrol.

The experimental studies used fuels with varying

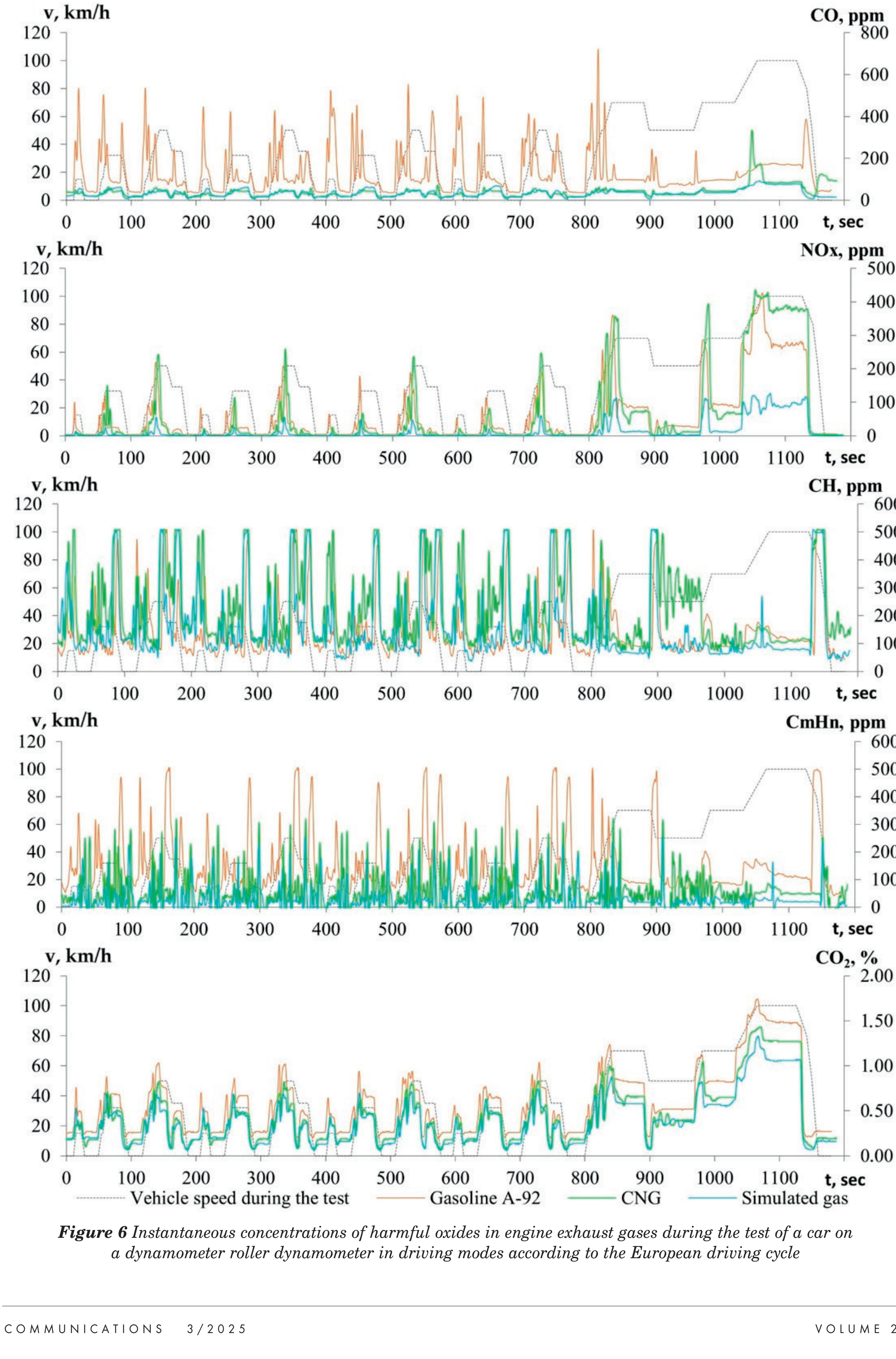


Figure 6 *Instantaneous concentrations of harmful oxides in engine exhaust gases during the test of a car on a dynamometer roller dynamometer in driving modes according to the European driving cycle*

***Table 4** Mass emissions of harmful substances during the testing of a car on a dynamometer roller bench in driving modes according to the European driving cycle*

Mass emissions of harmful substances	A-92 petrol	CNG	Simulated gas
G_{CO}, g/km	2.7743	0.7379	0.5040
G_{CO2}, g/km	180.8	127.3	106.4
G_{CH}, g/km	1.5051	2.7941	1.7749
G_{NOx}, g/km	1.7855	1.7580	0.4269
G_{CmHn}, g/km	1.4502	0.7352	0.3334
$G_{\Sigma CO}$, con. g/km	80.741	75.315	19.103

specific heat of combustion. The quality of heat use was assessed by fuel consumption in energy equivalent (MJ/cycle). The results show that when the engine is powered by CNG (46 MJ/kg), there is a 9.1% reduction in fuel consumption in energy equivalent. Similarly, when powered by simulated gas (43.2 MJ/kg), there is an 11.8% reduction compared to A-92 petrol (44 MJ/kg), which was used in the driving cycle.

Based on the obtained concentrations, the mass emissions of individual components and total mass emissions reduced to CO were calculated (Table 4).

The use of simulated gas in cars improves their environmental performance compared to A-92 petrol. Specifically, when powered by CNG, carbon monoxide (CO) emissions are 73.4% lower, and nitrogen oxides (NO_x) emissions are 1.5% lower. When powered by simulated gas, CO emissions are 81.8% lower, and NO_x emissions are 76.1% lower. Additionally, CO_2 emissions are 29.6% lower when powered by CNG and 41% lower when powered by simulated gas. When powered by simulated gas, emissions are 6% lower compared to non-simulated gas. Additionally, non-methane hydrocarbons (H_{CmHn}) are 49.3% lower with CNG and 77% lower with simulated gas. However, total hydrocarbons (H_{CH}) increase by 46.1% with CNG and by 15.2% with simulated gas.

The facility's environmental assessment calculates the total mass emissions reduced to CO using mass emissions and their relative aggressiveness coefficients (CO = 1, NO_x = 41.1, CH = 3.16).

Table 4 displays the total mass emissions reduced to CO. When powered by CNG, the total mass emissions of harmful substances in exhaust gases, reduced to CO, in the European driving cycle are reduced by 1.1% compared to A-92 petrol. Similarly, when powered by simulated gas, the emissions are reduced by 76.3% compared to A-92 petrol.

The novelty of this study can be summarised in the following aspects:

The study confirms a significant reduction in emissions of harmful substances in exhaust gases when using a mixture of biogas and compressed natural gas compared to traditional petrol.

The results of the study show an improvement in fuel efficiency, which is important for reducing the fuel costs and increasing the energy efficiency of vehicles.

The study's results suggest that a combination of biogas and compressed natural gas could be implemented for everyday use, contributing to the adoption of renewable energy sources in the transport sector.

These aspects underline the significant potential and importance of the research in the context of energy sustainability, environmental safety and innovation in the transportation.

4 Conclusions and prospects for further research

The results of the studies show that the use of mixed fuel (which was replaced by a similar simulated gas during the tests) reduces the concentrations and emissions of harmful substances in the exhaust gases.

Tests of a car equipped with a spark ignition engine, converted to run on CNG, show that mass emissions of harmful substances with exhaust gases, namely CO, G_{CmHn}, and G_{NOx}, decrease when the engine is powered by CNG, and simulated gas, compared to A-92 petrol. However, G_{CH} increases.

The test results showed that when the engine is powered by CNG, the total mass emissions of harmful substances reduced to CO, in the European driving cycle, are reduced by 1.1%, and by simulated gas - by 76.3%, compared to A-92 petrol.

The use of biogas as a motor fuel for car engines can reduce energy dependence on oil fuels and emissions of harmful substances in exhaust gases. Additionally, biogas can be used as an additive to compressed natural gas, eliminating the need for additional resources for purification. The study results can serve as a scientific foundation for developing policies and regulatory standards for alternative fuel use in vehicles. This can aid in transitioning towards a more sustainable transportation system.

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Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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