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Comparison of Selected Technologies to Improve the Quality of Exhaust Gases from Landfill Gas Combustion

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Abstract: This paper presents the results of research on exhaust gases from landfill gas combustion. The measurements were carried out in a reclaimed landfill in Kiełcz (a passive degassing system based on gas flare). The research concerned the effects of selected exhaust gas purification technologies (platinum catalyzer on a ceramic carrier, molecular sieve, copper(II) oxide) for the quality of exhaust gases, and their particulate matter content. This paper aims to indicate which catalytic systems are most suitable for this gas type and their most efficient positioning in the flue gas duct. Due to increasingly stringent emission standards, the outcomes presented in this article could be helpful for landfill owners who wish to avoid paying fines for not complying with applicable limits. The measurements were carried out using a flue gas analyser, a particulate matter analyser fitted with a probe, and four thermocouples connected with the data recorder. The research outcomes determined the percentage reduction of pollutant emissions into the atmosphere (CO, NO_x, and particulate matter) using catalysts. The potential benefits of using catalysts in landfill gas combustion systems due to their operating temperature ranges are discussed.

Keywords: combustion; landfill gas; exhaust gases; catalyst

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1. Introduction

Municipal waste, which in European countries is neutralised by depositing in landfills, can be transformed into a system whose main task will be to produce landfill gas from the waste therein [1,2]. Instead of escaping into the air, landfill gas can be captured, converted, and used as a renewable energy resource, but under one condition—monitoring and analyses of harmful substances released to the atmosphere. As proved [3], there are pollutions from landfill gas combustion, that negatively affect the surrounding environment.

A massive number of organizations, including the European Environmental Agency (EEA), have shown that air pollution is the greatest threat to human life and that appropriate restrictions introduced to reduce emissions of harmful substances will contribute to reducing the occurrence of this phenomenon [4]. The harmful substances that are emitted during combustion include mainly PM2.5 and PM10 dust, ozone (O_3), carbon monoxide (CO), nitrogen oxides (NO_x), sulfur dioxide (SO_2), benzopyrene (BaP), which is part of groups of polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/F), (polycyclic aromatic hydrocarbons (PAHs) and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) [4–6]. Human interaction with each of these substances leads to several unwanted and often even serious consequences. For example, PM2.5 and 10 are very dangerous to human and animal health as they are the main causes of pulmonary arterial hypertension (PAH). Suspended dusts are highly carcinogenic, and it is, therefore, necessary to reduce their emissions in any fuel combustion system. Carbon monoxide (CO) and nitrogen oxides (NO_x) adversely affect the proper functioning of the circulatory system (mainly carbon monoxide), the respiratory system (mainly NO_x), and concentration disorders [7–9]. Ozone (O_3) causes adverse effects, such as inflammation of the eyes, respiratory diseases, skin diseases, etc. [10]. In addition, sulfur dioxide (SO_2) also adversely affects humans, causing the symptoms of dyspnea, cough, cardiovascular diseases, and respiratory system diseases, mainly asthma [11]. Other extremely harmful compounds are benzopyrene (BaP), polychlorinated dibenzo-p-dioxins, and dibenzofurans (PCDD/Fs). Benzopyrene causes respiratory diseases, the formation of lung, skin, and bladder cancers, and contributes to ADHD and the cognitive process. In contrast, polychlorinated dibenzo-p-dioxins has a tremendous impact on the endocrine system and affects the prenatal development of women in pregnancy. It also affects the immune system, which leads to more infections [12–14].

The EU Emissions Trading System (EU ETS) is one of the critical elements for improving the environment, which covers ~40% of all greenhouse gases (GHG) emissions within the European Economic Area. This task is optimally (and above all cost-effectively) to reduce greenhouse gas emissions to the environment. It causes a continuous reduction of the limit reducing the total emission [15]. All restrictions aim to transition to a climate-neutral economy, which, according to the European Commission, will be met by 2050 [16]. In addition, according to the EEA analysis, the use of appropriate techniques and continuous implementation of new targets will be able to reduce sulfur dioxide (SO_2) emissions by 91%, PM dust by 82%, and nitrogen oxides (NO_x) by up to 79% [17]. For this reason, the search for a solution to this problem was started and the influence of catalytic additives during the combustion process began to be investigated.

Catalytic additives are a specific group of substances that, during combustion, are designed to increase and improve the combustion process of fuels (including landfill gas) while increasing the fuel oxidation processes along with incomplete combustion products. Depending on the catalyst used, it is possible to reduce the specific group of harmful substances. For example, the use of a catalyst may reduce acid rain, greenhouse gas emissions, and even the elimination of carcinogenic, mutagenic, and toxic compounds (by supporting the incomplete combustion process) [18]. Catalytic additive based on copper (Cu) and manganese (Mn) oxides, where the carrier is aluminum oxide (Al_2O_2) (however, the transmitters can also be ZrO_2 and TiO_2), showed a positive effect on reducing the emission of carbon monoxide (CO) and particulate matter in the combustion of biomass fuels. An additional advantage of this catalyst is its very low cost [19].

Additionally, the catalyst based on sulfur and other compounds blocks the process of dioxane (PCDD/F) synthesis by converting copper chloride ($CuCl_2$) into a milder copper sulfate catalyst ($CuSO_4^-$), which will reduce the formation of dioxins by up to 70%, and even with the use of additional compounds such as urea or ammonia reduce the emitted nitrogen oxides (NO_x) [20,21]. However, the number of catalytic additives cannot be exaggerated because if the concentration of specific compounds is too high (e.g., copper salt and sodium chloride), it will increase the emission of persistent organic compounds (POPs) [18]. In addition, during the research in 2021 at the Wroclaw University of Environmental and Life Sciences 5 different catalytic compounds (TiO_2, MnO_2, $Cu(NO_3)_2{\cdot}3H_2O$, 8% K_2PtCl_2 solution, and 32% urea water solution) were taken into account, using sodium aluminosilicate as a carrier of the active substance. The study showed that the use of catalytic additives in the combustion of sunflower husk pellets led to a reduction in the emission of harmful substances, increase of the combustion temperature, which contributed to the combustion of carbon monoxide (CO), i.e., to reduce the share of incomplete combustion in the boiler, and contributes to the amount of fuel burned [22]. Nevertheless, due to several favorable properties, catalytic additives, such as reducing the emission of harmful pollutants (CO, NO_x, SO_2, PM, PAH, PCDD/F), positively affect the combustion efficiency and extend the effective operation of plant components, such as the boiler [21].

However, for the catalytic additives to be well selected, according to specific variables (in this case landfill gas composition), attention must be paid to the waste management itself in landfills and an appropriate system for managing and controlling this gas. The creation of various degassing systems of landfills prevents the uncontrolled release of gas, mainly methane (CH_4) and pollutants falling into groundwater and greenhouse gas emissions. The use of such systems has a preventive effect and is able to generate landfill gas that can be used as an energy source [23]. Depending on the intended use, there are two systems divisions according to the intended use: utilization of LFG by burning it in a flare, generation energy for utility purposes (e.g., a gas engine conjugated with an electricity generator, a gas boiler). A typical landfill gas collection and control system (GCCS) has elements such as extraction wells, piping system for transporting the produced gas fuel, condensate management system, blower, and flare system, gas engine, generator, monitoring devices [23]. Extraction wells can be divided into two types: vertical and horizontal wells. The main difference between the two types is that vertical wells apply when, at a given site of landfill gas production, the plant will not receive new portions of waste for at least one year, but horizontal wells are used when the plant is systematically replenished with new waste [23–26]. The next step will be a blower and monitoring system consisting of sensors, temperature measurements, biogas (volumetric flow meter), and biogas composition (mainly methane). This is especially important to gather the necessary information about greenhouse gases (GHG) emissions, odor, and other problems affecting fauna and flora. After completing a series of measurements, the blower "blows" the gas into the flares or the gas engine with a generator.

Flares can be divided into two types: open flares and enclosed flares. The main difference is the price and simplicity of construction. Open flare is a simpler and cheaper solution that will ensure up to 98% efficiency in destroying harmful compounds. Therefore, it is estimated that the combustion efficiency of open flares is 50%, and the efficiency of closed flares is 90%. In most cases, the flare system is connected with energy use, for example, by a regular gas engine connected to a generator. The most commonly used engines are those that burn gas with low technical parameters. Then, the produced electricity can be left unchanged or converted into heat [23,25,27].

The landfill gas consists of two main compounds: methane (CH_4) and carbon dioxide (CO_2), the percentages of which are 30–60% for methane and 2–50% for carbon dioxide. It also includes small amounts of nitrogen, oxygen, ammonia, sulfides, hydrogen, carbon monoxide, and nonmethane organic compounds (NMOCs) such as trichloroethylene, benzene, and vinyl chloride. The emission of methane, which is the main component of landfill gas, significantly contributes to the deepening of the greenhouse effect [28]. The important benefit of its combustion is the conversion of methane into carbon dioxide (methane is a potent greenhouse gas 28 to 36 times more effective than CO_2 at trapping heat in the atmosphere over a 100-year period) [29]. Nonetheless, to reduce the number of harmful substances, several methods can be used, such as the SCR method (NO_x reduction and oxidation of CO and CH), SNCR method (ammonia or urea reduce NO_x to nitrogen and water), lime method (use of calcium carbonate, which lowers the flue gas temperature and then absorbs acid pollutants), reduction with activated carbon (reduction all impurities, crystallization of sulfur dioxide to sulfuric acid). Part of landfills commonly uses landfill gas treatment technologies which reduce hydrogen sulfide and/or volatile organic compound (VOC) content. In the case of SO_2 reduction, commonly used is the SulfaTreat system, which consists of large vessels filled with SulfaTreat media (iron-based granulate), through which LFG flows. The media chemically reacts with H_2S to form stable iron pyrite. In comparison, VOC removal systems are based on the principle of adsorption and have been widely and successfully deployed over a wide range of conditions. They employ activated carbon, which adsorbs VOCs onto its surface until the media's adsorption capacity is exhausted [30–33]. The interest in this topic constantly increases, which may be the beginning of new scientific discoveries.

Catalytic flue gas cleaning methods are increasingly used in fossil fuel heat and power generation systems. Until now, no one has dealt with the treatment of flue gases from landfill gas combustion. A certain innovation is the use of permanent catalytic systems in a landfill gas flare, thus reducing the number of pollutants in the exhaust gases. It is known that the fumes from the combustion of landfill gas contain high concentrations of pollutants. In the areas surrounding a landfill with a passive degassing system, air quality is usually worse. Referring to the scale of the country or the entire European Union, the problem of emissions from landfill gas combustion is significant [34,35].

For this reason, this work will be devoted to comparing the effect of selected catalytic systems on the concentration of individual substances in the exhaust gases from landfill gas combustion. The structure of the paper is as follows: Section 1 includes information on air pollutants and their impacts on health, type of catalysts, degassing systems of landfills. Section 2 presents the methodology for measuring exhaust gases' quality (NO_x, CO particulate matter content), and its temperature. Section 3 shows the effects of using selected exhaust gas purification technologies (platinum catalyzer on a ceramic carrier, molecular sieve, copper(II) oxide) for the landfill gas combustion efficiency. Section 4 provides a summary and a discussion on the research outcomes. Section 5 contains conclusions resulting from the conducted analysis and presents further plans of the research team in this regard.

2. Materials and Methods

2.1. Description of the Test Stand

Measurements were made at the landfill of reclaimed municipal waste in Kiełcz, Nowa Sól commune, Nowa Sól county, Lubuskie voivodship. The deposit of the landfill consisted of municipal waste. It was selected due to the fact that it was one of the closest landfills to Wrocław after a full reclamation process, equipped with a passive degassing system. Detailed information on the parameters of the landfill is presented in Table 1.

Table 1. Parameters of the reclaimed municipal waste landfill in Kiełcz.

Parameter	Unit	Value
Total capacity	m^3	1,800,000
Total capacity within the crown of the landfill	m^3	69,000
Mass of deposited waste	Mg	17,000

The landfill has a PEHD foil seal and a groundwater monitoring system. The landfill was closed and rehabilitated in 2017. Due to the fact that the landfill has been closed and rehabilitated, its morphology has not changed. The stabilised morphology ensures that the gas composition does not undergo major changes. The reclaimed landfill consists of three plots with a passive degassing system (with a torch to burn the landfill gas). Each accommodation unit has 20 gas collecting wells. All the wells are connected by a gas collector, which supplies gas to the buffer tank. After reaching the appropriate pressure in the tank, the gas is burned in a single-torch flare. Figure 1 is an aerial photograph of the reclaimed municipal waste landfill site in Kiełcz.

The measuring stand was a gas torch. It consisted of a chimney with measuring openings, a gas burner, a fuel-air mixture regulation system, gas pipes, and a stand. The lances of the exhaust gas analyser, dust meter, and temperature sensors were placed in the measuring openings. The torch was connected to the degassing system of the landfill through a special gas connection.

Measurements were made in 5 positions. The exhaust gas composition, dust, and temperature were measured at each position. The location of the measurement openings in the gas flare is shown in Table 2.

Table 2. Description of measurement points.

Position	Value
1	25 cm below the top of the torch
2	40 cm below the top of the torch
3	55 cm below the top of the torch
4	70 cm below the top of the torch
5	85 cm below the top of the torch

The test stand where the landfill gas was burnt is shown in Figure 2.

Figure 1. Reclaimed municipal waste landfill in Kiełcz.

Figure 2. Measuring stand at waste landfill in Kiełcz.

Measurement points have been shown graphically in Figure 3.

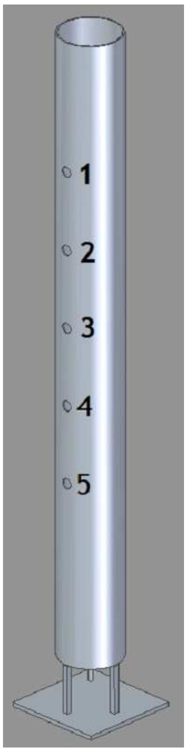

Figure 3. Measurement points located on the torch.

2.2. Applied Catalytic Additives

Three types of catalysts were used for the research: ceramic platinum catalyst, molecular sieve, and molecular sieve covered with a layer of copper(II) oxide (CuO). They are shown in Figure 4.

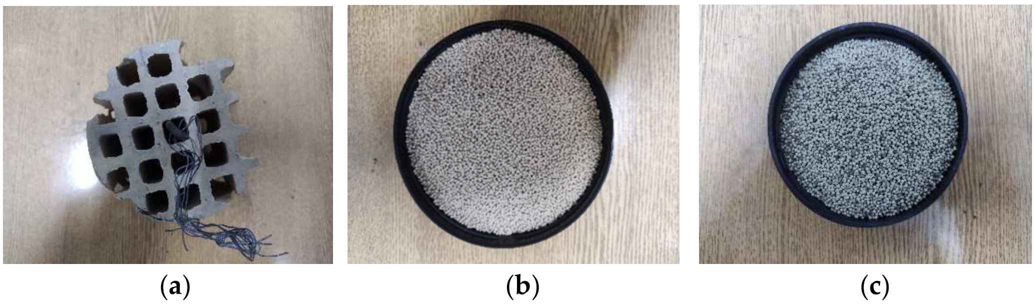

Figure 4. Catalysts used in the measurements: (**a**) platinum catalyst on ceramic carrier; (**b**) molecular sieve; (**c**) molecular sieve with copper(II) oxide.

2.3. Measurement of the Exhaust Gases Quality

The measurement of the exhaust gases quality (using ISO and EN standards shown in Table 3) was performed using the Testo 350 analyser. Detection of individual compounds contained in the exhaust gas is performed by a photochemical method. The recording of the exhaust gas composition began after the combustion process had stabilised and the analyser was calibrated. The analyser lance was placed successively in the measuring openings upstream and downstream of the catalyst. The values were recorded with the frequency of 1 s for 1 h of uninterrupted operation of the gas burner.

The technical specification of the analyser is summarised in the Table 3.

Table 3. Technical specification of the Testo 350 flue gas analyser.

Component	Measurement Method	Range	Precision	Compl. with Standards
O_2	paramagnetic	0–25%	±0.1% abs. or 3% rel.	EN 14789; OTM-13
CO	chemiluminescence	0–10,000 ppm	±3 ppm abs. or 3% rel.	EN 15058; METHOD 10
CO_2	chemiluminescence	0–25%	±0.03% abs. or 3% rel.	ISO 12039; OTM-13
NO_x	chemiluminescence	0–1000 ppm	±3 ppm abs. or 3% rel.	EN14792
SO_2	chemiluminescence	0–800 ppm	±5 ppm abs. or 5% rel.	EN14793

2.4. Measurement of the Particulate Matter Content in Exhaust Gases

The solids content of the exhaust gas was measured with the Testo 380 particulate matter analyser. The content of the sum of suspended dusts in the flue gas was determined, disregarding the division into individual fractions. It has been carried out following the standard PN-EN. The analyser counts the amount of PM in the exhaust gas through infrared detection. Before starting the measurement, the analyser was conditioned. The measurement was started when the combustion process stabilised. Dust registration lasted 1 h of uninterrupted operation of the gas burner. The values were recorded with a frequency of 1 s.

The technical data of the Testo 380 analyser have been shown in the Table 4.

Table 4. Technical data of the Testo 380 analyser.

Component	Measurement Method	Range	Precision	Compl. with Standards
PM	NDIR	0–300 $mg \cdot m^{-3}$	±1 ppm abs. or 1% rel.	EN14842
CO_2	chemiluminescence	0–20%	±0.03% abs. or 3% rel.	ISO 12039
O_2	paramagnetic	0–22%	±0.1% abs. or 3% rel.	EN 14789

2.5. Measurement of the Exhaust Gases Temperature

The exhaust gases temperature was measured following the standard PN-EN using the APAR AR205 recorder to which four K-type thermocouples were connected. The first thermocouple was placed above the burner and recorded the flame temperature, the second one in front of the catalyst, the third one after the catalyst, and the fourth one at the fumes outlet from the torch. The temperature was recorded every second during the entire period of the torch operation. Based on the recorded temperature parameters, average values were determined. The results took into account the mean measurement error, which was ±1.5 °C.

3. Results

The research was conducted to compare the effect of selected catalytic systems on the concentration of individual substances in the exhaust gases from landfill gas combustion. The temperature distribution inside the gas flare was determined to find the optimal place for the catalyst to work and compare the operating temperature's influence on its efficiency. The temperature distribution and the pollutant concentration values for all catalyst variants and its location in the flare were compared. This approach allowed for selecting the optimal solution in terms of reducing pollutants in the exhaust gases. The measurement results of exhaust gases quality and particular matter content were converted into a 3% oxygen content in the exhaust gases.

3.1. Exhaust Gases Temperature

Figure 5 shows the average temperature that was recorded in every measurement point located in the gas flare.

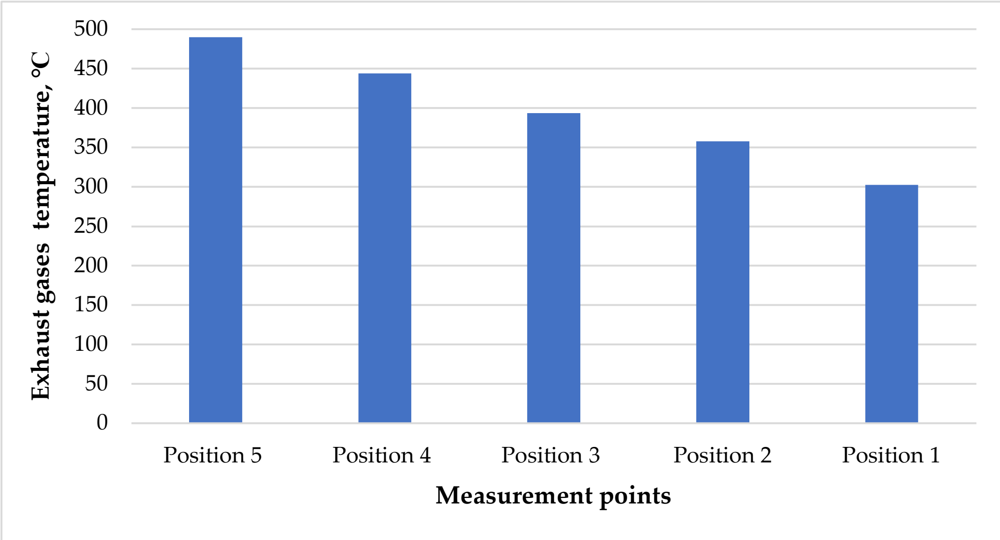

Figure 5. Temperature of the exhaust gases at various measurement points.

It can be observed that the temperature during the combustion of landfill gas fluctuated in the range of 300–490 °C. Considering the temperature range in which simple catalytic systems work (50–650 °C), their use in a gas flare was justified.

3.2. Exhaust Gases Quality

Figure 6 presents the dependence of CO concentration in the exhaust gases on the type of catalyst and its position in the flue gas duct.

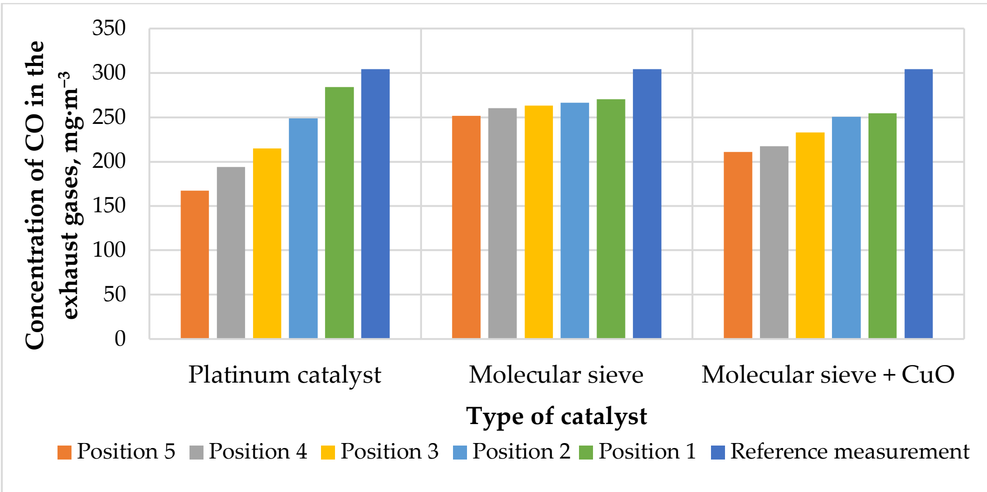

Figure 6. CO concentration in exhaust gases depending on the type and location of the catalyst.

The concentration of harmful carbon monoxide in the exhaust gases, where no catalyst was used, was on average 304 mg·m^{-3}. Depending on the type of catalyst used and the place of its installation, the CO concentration in the exhaust gases decreased to a different extent. The use of a platinum catalyst reduced the concentration of CO in the exhaust gases in the range of 20–137 mg·m^{-3}. Using a catalyst in the form of a molecular sieve bed, the average concentration of CO in the exhaust gases was reduced in the range of 34–53 mg·m^{-3}. After placing a bed of molecular sieve in the torch, on the surface of which CuO had been deposited, the concentration of carbon monoxide was reduced by an average of 49–93 mg·m^{-3}.

The concentration of NO_x in the exhaust gases' dependence on the type of catalyst and its position in the flue gas duct has been shown in Figure 7.

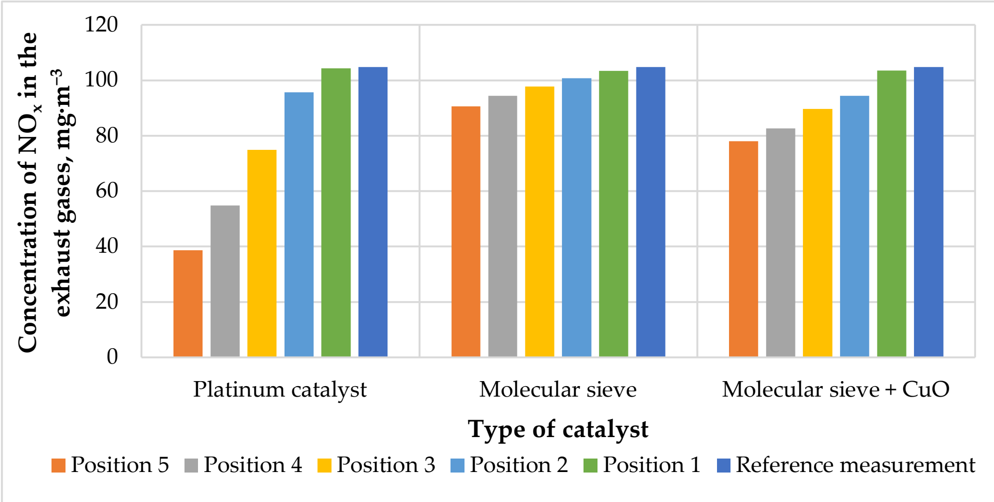

Figure 7. NO_x concentration in exhaust gases depending on the type and location of the catalyst.

Nitrogen oxides are an undesirable component of exhaust gases that significantly deteriorates the quality of the environment. During the combustion of landfill gas in a flare without the installed catalyst, the emission of these compounds was on average 105 mg·m^{-3}. The use of catalysts in each configuration reduced the NO_x concentration in the exhaust gases. The use of a platinum catalyst reduced the concentration of NO_x in the range of 0.4–66 mg·m^{-3}. After installing the catalytic bed in the form of a molecular sieve, the average NO_x concentration in the exhaust gases decreased by an average of 1–14 mg·m^{-3}. The catalyst in the form of a molecular sieve with a layer of CuO reduced the concentration of NO_x in the exhaust gases in the range of 1–27 mg·m^{-3}.

3.3. Particulate Matter Content in Exhaust Gases

Figure 8 shows the impact of catalyst type and its location in the flue gas duct on the concentration of particular matter (PM) in the exhaust gases.

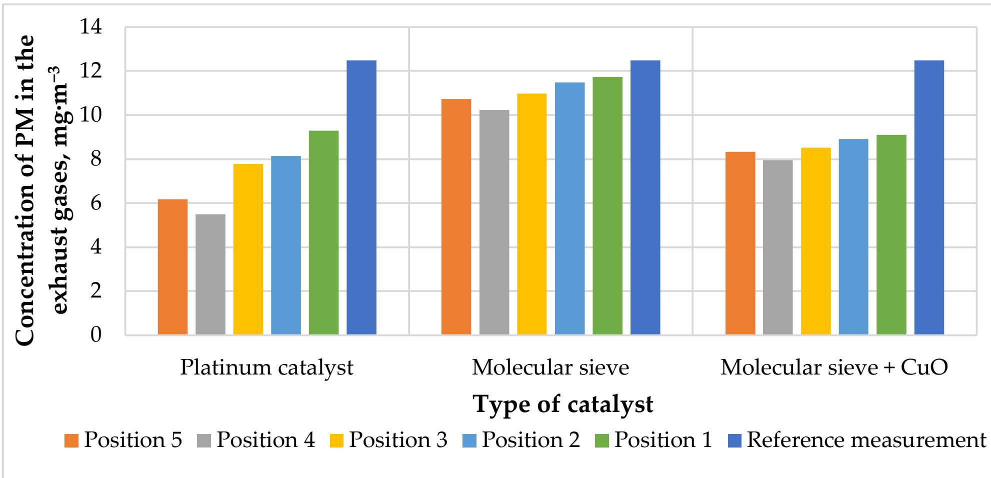

Figure 8. PM concentration in exhaust gases depending on the type and location of the catalyst.

During the combustion of landfill gas in a flare without the use of catalysts, the average PM concentration was 12.5 mg·m^{-3}. After installing the platinum catalyst, the PM concentration in the exhaust gases decreased on average in the range of 3–4.7 mg·m^{-3}. It can be seen that the greatest degree of PM reduction was recorded in the fourth position of the catalyst location. The flare with the installed catalyst in the form of a molecular sieve bed emitted exhaust gases with a lower PM concentration in the range of 0.7–2.2 mg·m^{-3}.

A catalyst based on a molecular sieve with a layer of CuO caused a decrease in PM concentration in the exhaust gases in the range of 3.3–4.5 mg·m^{-3}.

4. Discussion

Due to the unique nature of the research, the chemical composition of the landfill gas, and the catalysts used, it is not possible to compare the results of the research with the results of similar studies. However, the general benefits of using a ceramic platinum catalyst, molecular sieve, and molecular sieve covered with a layer of copper(II) oxide (CuO) used in the research have also been reported by other researchers. The main finding from the research was that the most expensive catalyzer (platinum one) was the most efficient in terms of reducing NO_x and CO. It allows to reduce dust from gases (PM) by 50% and NO_x by almost 60%. Also, the efficiency in CO reduction (45% respectively) is significant. The research showed also a positive influence on combining a bed of molecular sieve in the torch with CuO which had been deposited on the surface, and the concentration of emissions was reduced by 15–30%, depending strongly on the catalyzer position. The emission reduction is lower than by platinum catalyst, but the price is much lower as well.

The position of the catalyzer was found to be one of the most important factors determining the reduction. The results indicated that the optimal catalyzer position is a function of the gas temperature and that graphs of emissions and gas temperature should be used to determine the position of the catalyzer used those results in the lowest emission (see Figures 6–8). For the selected position the optimal temperature for chosen catalyzer can be found in Figure 5. Interestingly, for most of the measurements in this study, the optimal temperature was found to be 440 °C.

The application of results described in the article could significantly lower emissions from landfill gas combustion, which are commonly used in the European Union countries. On the scale of the country and the entire European Union, the use of such a solution would improve the quality of atmospheric air.

5. Conclusions

The approach presented in the manuscript is novel. So far, no research team has undertaken to investigate the effect of catalytic additives on the quality of exhaust gases from landfill gas combustion. Only landfill gas treatment methods were analysed.

Similar analyses were performed for catalytic additives for combustion with wood pellets [36], sunflower husk [22], or for using urea to reduce NO_x emissions [37] from low power biomass boilers. Catalytic additives were efficient in reducing emissions, and profitability of their use has risen, resulting from the reduction of fuel consumption as a result of increased combustion efficiency. The urea could be effective as well [38] but the emission reduction effect was bound with the optimal temperature in the combustion chamber. The control of the temperature was complicated in the landfill gas combustion installation, and it was the main reason to use other technologies (described in the paper) for cleaning the exhaust gas. There was also research for the effect of fuel type and active substance addition on exhaust gas emissions from vehicles powered by a spark ignition engine [38]. Added emulsions of TiO_2, TiO_2-CuO, and TiO_2-Pt nanoparticles have decreased the NO_x, CO, particular matter, and hydrocarbons emissions. That research was also an argument to use catalytic additives for landfill gas combustion treatment.

The findings in other articles show that catalytic additives are a proper technology for exhaust gas cleaning, including landfill gas. The main scientific problem is finding the proper balance of catalytic compounds to combine high reduction efficiency with the low cost of the chosen substance. The article was intended to help find the proper catalyst for landfill gas combustion.

This research makes it possible to implement this type of solution in municipal landfills with a passive degassing system. However, there are some limitations in the application of catalytic additives. The active substance of the catalytic system is characterised by the most effective operation in a specific temperature window. A too low or high temperature of the exhaust gas may deteriorate its efficiency. It is also worth noting that catalytic systems do not affect the efficiency of thermal conversion of landfill gas, as they are only intended to improve the quality of combustion products. The paper also creates a space for further research as research on the viability of catalytic systems, the effect of hydrogen sulfide on the degradation of the active surface of the catalyst, expansion of the base of catalytic additives with new active substances (the willingness to increase the efficiency of exhaust gas cleaning). The research team also plans to investigate the impact of catalytic systems on the quality of exhaust gases in other types of landfills, including those with an active degassing system.

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