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ORIGINAL PAPER

Environmental aspect of waste to energy installation: quality of waste generated by technology

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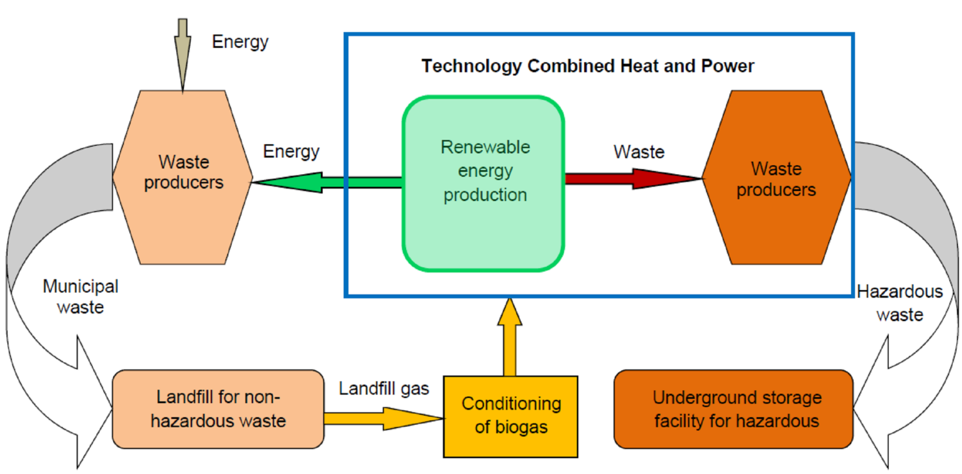
Abstract

Production of green energy from landfill gas in a cogeneration system is considered optimal in terms of the use of local renewable energy resources, which in the production process generates hazardous waste. Yet, the production of renewable energy is not without environmental effects. The objective of the work was to analyse the operation of the installation for biogas energy generated at a municipal waste landfill and to carry out tests on the waste (mineral deposit) from a heat exchanger generated in the exhaust manifold of a gas engine of the cogeneration unit. The objective of the work was to test waste (mineral deposit) from the heat exchanger in the exhaust manifold of the gas engine of the cogeneration unit. The conducted research constitutes a significant contribution to the identification and classification of hazardous waste generated in a cogeneration unit powered by landfill gas. The results of the research showed that the waste contains high concentrations of ecotoxic elements, i.e. heavy metals (arsenic, chromium, copper, nickel, zinc), molybdenum, antimony, sulphates, which have carcinogenic activity. Pioneering research performed as part of the leaching test showed that the permissible concentrations were exceeded for 11 parameters in the case of storage in a hazardous waste landfill, and for 6 parameters in the case of storage in an underground hazardous waste landfill. The research and analyses performed in this work will serve as input material for defining the methods of hazardous waste disposal from the cogeneration unit.

Graphical abstract



Circular economy in the operation of a waste landfill

Keywords Cogeneration · Waste-to-energy · Heavy metals · Hazardous waste · Waste procedures

Abbreviations

ANC Acid neutralization capacity (mmol dm^{-3})
CHP Combined heat and power

Extended author information available on the last page of the article

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DOC Dissolved organic carbon (mg kg^{-1} dry weight)
GRI Global reporting initiative
LFG Landfill gas
LOI Loss on ignition (% dry weight)
TDS Total dissolved Solids (mg kg^{-1} dry weight)
TOC Total organic carbon (% dry weight)

Introduction

Waste management should comply with a hierarchy of approaches that give priority to prevention, recycling and recovery. Waste recovery is a process in which waste is reprocessed into products, materials or substances, and the recovered chemical elements with important properties are reused in industry and in its many branches. The law also allows the use of waste disposal by landfilling, but this form is a last resort, as it generates negative effects (Tomić et al. 2022; Act on waste 2012; Generowicz and Kulczycka 2020). Municipal landfills, in line with the definition stipulated in the construction law, are construction objects which, during their operation, directly or indirectly, have a negative impact on the natural environment or human health. Therefore, in this context, it is important to ensure a transparent supervision of a landfill in terms of the management of waste or sewage that are not dumped at the landfill, but are produced in the installations located on the site of the landfill (Gronba-Chyła et al. 2022; Anandha Kumar et al. 2023). This mainly involves emissions, wastes and the pollution of surface or ground waters effected by a leakage in the landfill insulation and uncontrolled outflow of leachate into water. Wastewater generated in the landfill has a varied composition, and it most frequently contains heavy metals and toxic compounds that pose a threat to living organisms (Yilmaz et al. 2021; Smol et al. 2017).

The tests of surface and ground waters carried out at sampling points located near the landfill often show exceedance of permissible values. Such a state may have a potential impact on the quality of water collected in water intakes operating within the water treatment plant in a given area. In such a situation, it is required to conduct a continuous monitoring of waters in the area of the landfill, along with the analysis of the results and methods used to counteract possible exceedances of permissible parameters (Ciuła et al. 2023a, b; Wysowska et al. 2022). Wastewater generated at the waste deposit site should be captured, monitored and managed depending on its parameters. The key issue in this regard is to follow an appropriate selection of wastewater treatment technology, which should meet specific legal requirements and the mutual arrangements, e.g. with the operator of the wastewater treatment plant. In the case of discharge of wastewater from the landfill to the sanitary sewer system, appropriate arrangements are required with the operator of the sewage treatment plant in terms of permissible parameters so that the processes taking place in the treatment plant are not disturbed (Bugajski et al. 2015; Koc-Jurczyk et al. 2021; Ishchenko et al. 2017). The quality and quantity of biogas produced in the landfill waste bed are influenced by humidity, which can be supplemented by recirculation of leachate. This method requires monitoring and control of leachates to optimize the parameters of the LFG intended for purification and energy use in the energy conversion process. (Zhou et al. 2012; Gaska et al. 2019). In the waste deposited at the landfill and subject to an appropriate compromisation, biological processes take place. Their type and intensity are closely related to the morphology of the stored waste, and in particular to the amount of the biodegradable fraction. In such a situation, the process brings about the generation of landfill gas (LFG), whereof the main components are methane and carbon dioxide (Delgado et al. 2022; den Boer et al. 2021). For proper management of landfill gas used for energy production, it is necessary to parameterize individual wells collecting gas and monitor the parameters. This aspect is crucial in the context of the potential formation of hazardous waste in the energy production process, especially during its purification and preparation of the fuel mixture (Kutsyi 2015).

Taking into account the fact that methane is one of greenhouse gases, its collection and controlled thermal disposal is recommended. The collection of gas as a renewable fuel is the main argument for its optimal energy use in order to reduce the negative impact of the landfill on individual components of the environment and to protect human health (Putna et al. 2022; Aracil et al. 2018; Mukawa et al. 2022). The technologies involving energy application of LFG used currently allow for the optimization of this process, taking into account the efficiency of the use of LFG chemical energy. One of the most commonly used methods involves cogeneration systems, which allow to generate electricity and heat in one technological process (de Souza et al. 2021; Ciuła et al. 2022; Van Caneghem et al. 2019).

Landfill gas obtained from a waste deposit contains impurities. Hence, it is necessary to reduce harmful compounds contained in landfill gas by building a gas treatment installation. For this purpose, devices that apply the best available technologies and construction materials should be used, together with the development of a construction waste segregation system, directly on the construction site (Milewski et al. 2012; Dimitriu 2014).

The selection of equipment for LFG purification should be preceded by laboratory tests, involving the composition of individual biogas impurities, which allows to select an appropriate purification technology. In recent years, dry technologies have become popular, based on the adsorption of pollutants, mainly sulphur and silicon compounds, by activated carbon (da Silva Van Tienen et al. 2021; Tansel and Surita 2019).

Such compounds, reduced during the combustion of biogas in the engine compartment, bring about the formation of deposits on individual parts of the engine, causing their intensive wear. The chemical composition of such deposits is mainly made of sulphur, silicon, calcium, aluminium, iron and zinc (Kowalski et al. 2021; Stanuch et al. 2020; Kowalski 2021). The deposition of mineral solids in the exhaust manifold containing the heat exchanger leads to reduction in the efficiency of heat exchange between the exhaust gas and the heating medium, which is a mixture of glycol (Ajhar et al. 2010; Tappen et al. 2017; Zięby and Gładysz 2017). When analysing the cumulative negative impact of a waste landfill as a construction object together with the accompanying technical infrastructure, one should also take into account the negative environmental impacts of cogeneration installations. This applies to the organized emission of pollutants to the air, resulting from the combustion of gas in the engine of the combined heat and power (CHP). It is a direct emission of gases and dust, reduced by the dust constituting the deposits (waste) developed in the exhaust manifold of a gas engine as well as in heat exchangers (Borsukiewicz-Gozdur 2010). The amount of dust emission is the resultant of the cogeneration installation power, fuel quality, installation operation time and the amount of the landfill gas stream burned per unit time in a gas engine (Regulation, 2015a). The installation of active degassing of a landfill interacting with the system of energy use of landfill gas in a cogeneration unit is also a place where waste is generated, including hazardous waste. Such wastes should be balanced in quantitative and qualitative terms and specified in relevant reports and documentation, including environmental fees. Transparency of their activities in relation to environmental and social issues is becoming increasingly important for building good relationships of organizations with both internal and external partners, including control and supervisory bodies. The research carried out in this work fills the gap in this area by identifying hazardous waste generated during the production of energy from a renewable source, along with their parameterization and methods of disposal. The results of these studies can be used when preparing reports and monitoring the environment, as part of the company's social and economic responsibility, including, for example, GRI Global Reporting Initiative reporting (Janusz-Szymańska et al. 2021).

The objective of the article is to present pioneering research on balancing the quantity and quality of waste generated as a result of the operation of 'waste to energy' installation—energy recovery from the installation that uses biogas from municipal waste. The novelty of the presented research is the quantitative and qualitative balance of waste, which will allow to define the net environmental impact of the installations in terms of renewable energy sources from the waste.

The absolute novelty of the work is the location and identification of hazardous waste and the fact that in the literature, not only Polish but also worldwide, there has been no study of deposits from a heat exchanger in the context of the threat it poses to the environment and humans. There are no cases in the literature of analysis of waste disposal methods from the heat exchanger of a CHP unit. There were also no reports on the monitoring of hazardous waste generated during the conversion of landfill gas in cogeneration units containing the analysed hazardous waste. Taking into account the above, the authors believe that they make a significant contribution to this field of science, demonstrating the novelty of an area of research that has not been the subject of interest and research so far.

Materials and methods

Research object

The object of the research involved a cogeneration unit for the production of electricity and heat in a container construction with an electric power of 345 kW and thermal power of 470 kW, which is operated in an active landfill for non-hazardous and inert wastes in Poland. The landfill has been operating for over 15 years, and due to the expansion of the degassing installation, a cogeneration unit was installed in March 2016, consisting of a piston engine powered by landfill gas, which drives a synchronous generator. Landfill gas with a variable methane content from 50 to 55%, and oxygen from 0 to 04%, is obtained from the landfill using an active degassing installation consisting of vertical and horizontal degassing wells. The gas subjected to the purification process in the carbon filter is used for energy generation in the CHP unit. In the year 2022 the degassing installation obtained 1,240,000 m^3 of landfill gas from the landfill, which was the source of electricity generation in the amount of 2,170 $MWh•year^{-1}$ and heat generation of 2,455 $MWh•year^{-1}$. The CHP installation from the first start-up at the landfill worked 15 thousand hours, and therefore, the first so-called minor overhaul of the engine was required, resulting from the operation schedule of this unit. The diagram of gas acquisition installation, its treatment and energy generation use in the cogeneration unit along with LFG monitoring and sampling points, is shown in Fig. 1.

The generation of electricity and heat in a cogeneration unit powered by landfill gas means not only green energy, but it also produces waste. It is related to physical, chemical and thermal processes taking place throughout the installation. In effect of burning landfill gas in a gas engine, the emission of dust contained in the exhaust gases takes place,

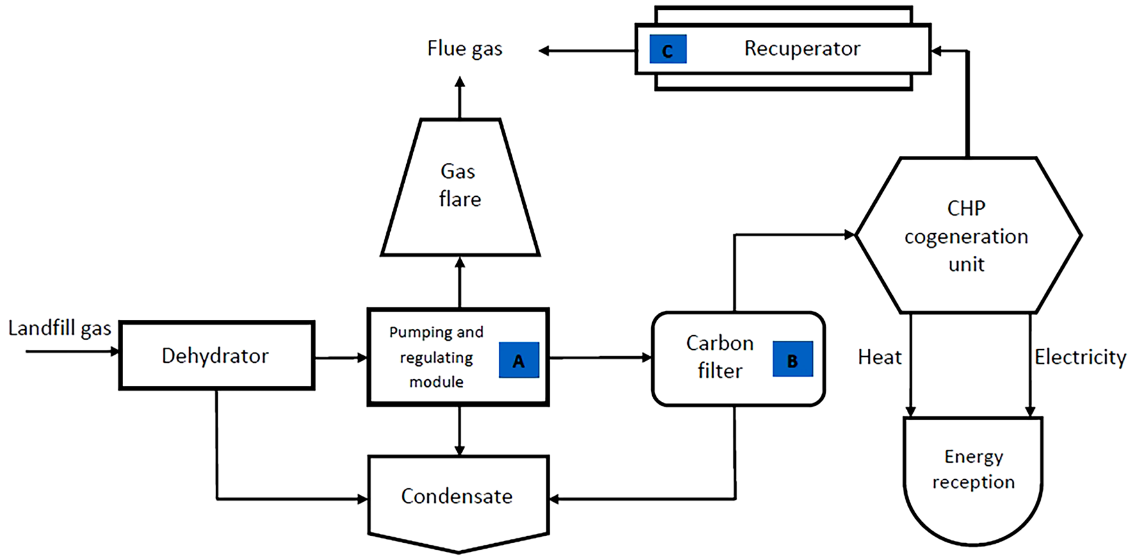


Fig. 1 Scheme of the installation for the energy use of landfill gas with monitoring and sampling points.

which is an organized emission into the air. However, not all of the dust contained in the exhaust gas is the emission to the air, since part of it is retained in the exhaust manifold containing the exhaust-glycol heat exchanger, which is a heat recovery block in the CHP unit. The planned repairs carried out in the cogeneration installation are intended to replace consumables, components and spare parts for the correct operation of the cogeneration unit. As part of these works, activities related to cleaning the heat exchanger located in the exhaust manifold of the gas engine from dust contamination are also planned. Mineral waste obtained in this way from the exhaust manifold containing the heat exchanger must be disposed of in a manner consistent with the requirements of the waste disposal law. This waste was classified as waste from exhaust gas treatment (deposit). Laboratory tests of the waste (deposit) were carried out in order to determine its chemical composition, which can ensure proper handling of this waste.

The research and analyses carried out in the work are aimed at determining the type of waste and the method of its disposal. This applies to the waste generated in the cogeneration unit in the heat exchanger, located in the exhaust manifold.

Methodology of research

Testing the quality of biogas in the degassing installation in the landfill (raw gas) was performed in the modular acquisition and control station (point A) and after its purification in the carbon filter (point B), before entering the gas engine. The content of methane, carbon dioxide, oxygen and hydrogen sulphide was tested. For this purpose, a portable Gas Data gas analyser model GFM 416 was used, which has a valid quality certificate ATEX II, and before the measurement it was calibrated with certified calibration gases based on the applicable procedure.

In order to determine the type of waste generated during energy generation in a cogeneration unit powered by landfill gas, three series of tests were planned. The dates of sampling for testing purposes were correlated with the dates of repairs of the cogeneration unit, reflecting the operational schedule. The first sample for testing was taken after 15 thousand of operating hours of the installation in July 2018, and the second sample was taken in June 2018, after working 30,000 motohours, while the third one was collected after 45 thousand motohours in May 2022. The dates of sampling for testing were correlated with the dates of renovation of the cogeneration unit. The service manual for this type of cogeneration units provides for the unit to be overhauled after 16,000 h of operation. hours, 30 thousand time. and 46 thousand hours. As part of these renovations, it is necessary to clean the heat exchanger system.

The test samples, being the waste (mineral deposit) effected by the combustion of landfill gas in the gas engine, were taken at point C from the heat exchanger in the exhaust-glycol system, which is installed in the exhaust manifold of the gas engine of the cogeneration unit (Fig. 1). For a cogeneration unit with an electric power of 345 kW, the amount of mineral deposits collected in the exhaust gas heat exchanger is variable and depends on the unit's operating time and the degree of utilization of the rated power. In the analysed case, this amount is approximately 11 kg per year, which is 0.005 g/kWh. The place of sampling for testing, and the mineral sediment being the subject of the tests are shown in Fig. 2.

Deposits formed inside the heat exchanger in the exhaust gas system were collected at the service company's headquarters during routine work after dismantling the heat exchanger from the cogeneration unit. In order to obtain the mineral deposit, impact techniques were used, which involve generating rhythmic blows transmitted by a pneumatic hammer tool to the metal casing of the heat exchanger at many of its defined points. After collecting the sediment, the heat exchanger was additionally cleaned with water under appropriate pressure.

The sample collected for testing was made up of wastes (deposits) with a granulation of 0.5 to 10 mm, and it was transferred to an accredited laboratory for further preparation for testing. Three series of tests were planned. The first series (sample No. 1) consisted in determining the basic elemental composition of the examined waste in micro-areas using a scanning electron microscope (SEM). During the second series of tests (sample 2), it was proposed to increase the scope (in relation to the test in the series 1) in order to determine the type of waste, including heavy metals characterized by the analysis of extractable metals (mainly cations), non-metallic inorganic parameters and physical parameters. In the third series of tests (sample 3, scope 1), the scope from the second series was repeated to compare the results of the tests obtained, to identify and determine the type of waste generated for its further management (Regulation 2015a). In addition, in the third series (sample 3, scope 2), a leaching test was planned for compliance testing with respect to the leaching of granular waste materials and sediments.

A test of the liquid to the solid-phase 10 l/kg was performed for materials with a particle size of less than 10 mm (with or without size reduction). The ratio of the liquid to the solid sample was 10:1. The leaching methods were used in accordance with Annex 5 to the Regulation of the Minister of Economy of 16 July 2016 on the admission of waste to be stored in landfills (Regulation 2020; Commission Regulation 2015).

Description of analytical methods

The waste obtained for the first series of tests was prepared in the laboratory in order to determine the morphology of its surface as well as the size and basic elemental composition of the tested waste in microareas, using a scanning electron microscope (SEM). For this purpose, 5 points were isolated on the surface of the waste. On the other hand, the waste collected for the second and third series of tests was crushed, ground and powdered. The tests were performed in an accredited laboratory.

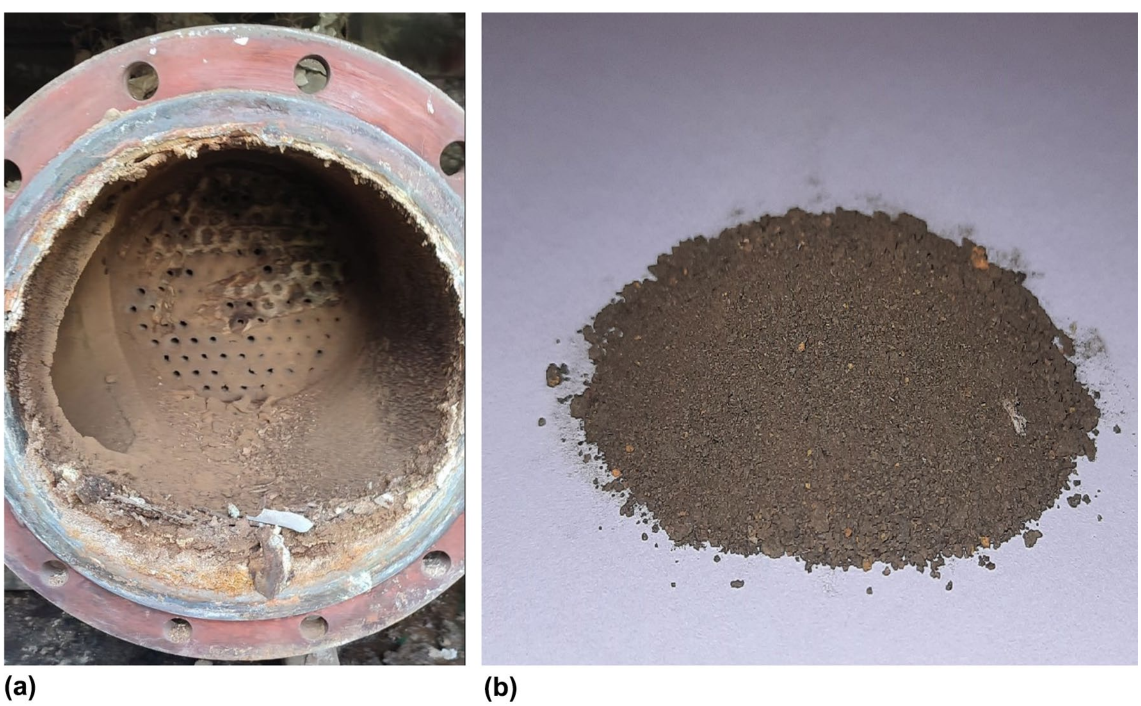


Fig. 2 Heat exchanger and waste. **a** Fragment of the heat exchanger in the exhaust gas manifold, **b** Sample of mineral deposit (waste) taken for testing

Research results

Test results of landfill gas

The control of the composition of landfill gas is essential for the correct operation of the cogeneration installation in terms of its reliability and efficiency of energy production. The daily adjustment of gas stream and its composition in the modular acquisition and control station is an important element of optimizing the use of gas resources in the landfill in terms of its acquisition and quality, Table 1.

The obtained results of testing of the basic parameters of landfill gas indicated correct energy values and a satisfactory reduction of hydrogen sulphide after its purification in a carbon filter. The high and stable content of methane in landfill gas indicates that in the landfill there is anaerobic decomposition of the organic fraction (methanogenic), resulting in the formation of methane, carbon dioxide and a decrease in nitrogen. The literature on the subject describes this condition as methanogenic phase IV—stable, appearing in waste deposits that are 2 to 10 years old after their deposition. This condition also proves that the landfill is operated correctly, especially in terms of waste compaction by specialized machines (Park et al. 2010).

Table 1 Parameters of landfill gas from the cogeneration installation

Parameter	Unit	Untreated gas	Purified gas
CH_4	%	54.2	54.1
CO_2	%	38.3	38.3
O_2	%	0.3	0.2
H_2S	ppm	247.8	31.8
Other	%	7.18	7.40

Preliminary studies of waste using a scanning microscope.

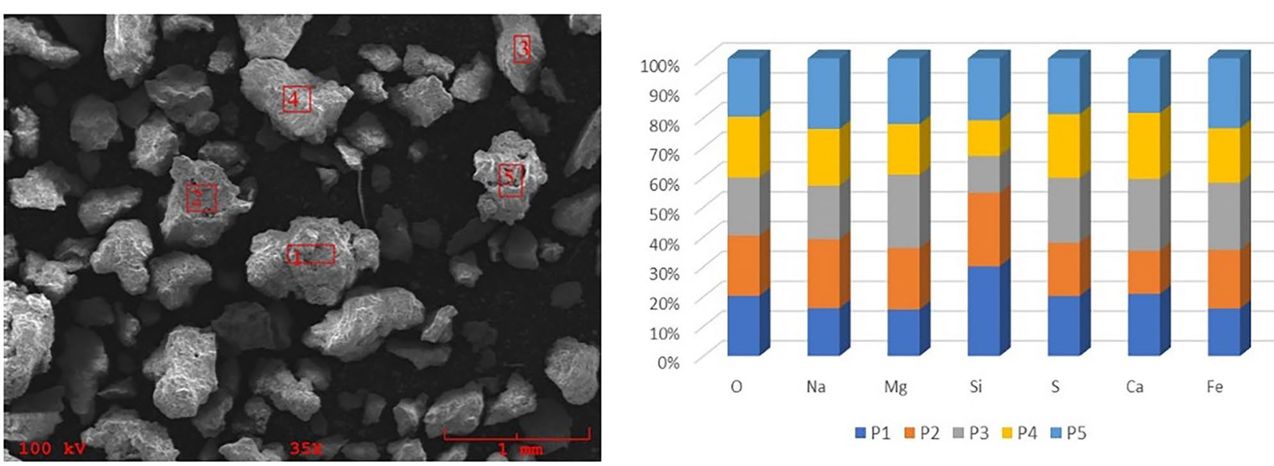
The first series of tests was performed after 15 thousand operating hours of the CHP unit, in order to determine the surface morphology and the size and basic elemental composition of the tested waste in micro-areas (points). Using a scanning electron microscope (SEM), a study was performed—a high-resolution image (Fig. 3a). Then, 5 points on the surface of the waste (P1–P5) were isolated, for which the elemental composition was determined. The test results are shown in Fig. 3b.

In the analysed waste, a high content of oxygen was reported (on average 75.17%), sulphur (on average 10.23%) and iron (on average 6.69). The waste also contained magnesium (on average 3.67%), silicon (on average 2.31%), sodium (on average 1.64%) and calcium (on average 0.31%).

Results of the solid waste fraction test—the second series of tests

As part of the second series of research, one test was planned (sample No. 2) with an increased scope, as compared to the first series, in order to determine the type of waste and its properties. The examination of the solid fraction for the content of chemical elements, including heavy metals, was characterized by the analysis of extractable metals (mainly cations), non-metallic inorganic parameters and physical parameters. The sample was collected after 30 thousand of operating hours of the CHP unit. The test results of the sample No. 2 are summarized in Table 2.

The results of the second series of tests for the sample 2 (Table 2) demonstrated the presence of high concentrations of heavy metals, including lead (1.8 mg·kg^{-1} d.m.),



(a) (b)

Fig. 3 Examination of waste in the first series: **a** Isolated points, **b** Test results involving the individual points

Table 2 Results of waste test in the second series for the sample No. 2

Parameter	Unit	Results	Measurement uncertainty
Extractable metals/mostly cations			
Sb	$mg \cdot kg^{-1}$ s.m	100	±19.9
As	$mg \cdot kg^{-1}$ s.m	34.6	±6.9
Ba	$mg \cdot kg^{-1}$ s.m	7.1	±1.41
Be	$mg \cdot kg^{-1}$ s.m	<0.010	–
Cr total	$mg \cdot kg^{-1}$ s.m	1141	±228
Sn	$mg \cdot kg^{-1}$ s.m	23.8	±4.7
Zn	$mg \cdot kg^{-1}$ s.m	124	±24.8
P	$mg \cdot kg^{-1}$ s.m	16.8	±3.4
Cd	$mg \cdot kg^{-1}$ s.m	<0.40	–
Co	$mg \cdot kg^{-1}$ s.m	31.2	±6.3
Li	$mg \cdot kg^{-1}$ s.m	<1.0	–
Mn	$mg \cdot kg^{-1}$ s.m	960	±192
Cu	$mg \cdot kg^{-1}$ s.m	86	±17.2
Mo	$mg \cdot kg^{-1}$ s.m	247	±95.7
Ni	$mg \cdot kg^{-1}$ s.m	1573	±314
Pb	$mg \cdot kg^{-1}$ s.m	1.8	±0.3
Hg	$mg \cdot kg^{-1}$ s.m	<0.20	–
Ag	$mg \cdot kg^{-1}$ s.m	<0.50	–
Sr	$mg \cdot kg^{-1}$ s.m	3.1	±0.62
Tl	$mg \cdot kg^{-1}$ s.m	<0.50	–
V	$mg \cdot kg^{-1}$ s.m	6.8	±1.36
Fe	$mg \cdot kg^{-1}$ s.m	19,693	±3900
Non-metal inorganic parameters			
Chlorides	mg/kg s.m	<40	–
F total inorganic	mg/kg s.m	210	±64
TOC	% dry mass	3	±0.45
SO_4^{2-}	% dry mass	8.6	±0.86
SO_3^{2-}	% dry mass	98,000	±14,700
Physical parameters			
Volume density	$g \cdot dm^3$	450	±137
Loss on roasting in 550 °C	% dry mass	33.1	±1.65
Dry mass in 105 °C	%	97	±5.85

zinc (124 $mg \cdot kg^{-1}$ d.m.), chromium (1141 $mg \cdot kg^{-1}$ d.m.), nickel (1573 $mg \cdot kg^{-1}$ d.m.), copper (86 $mg \cdot kg^{-1}$ d.m.), iron (19,693 $mg \cdot kg^{-1}$ d.m.) and arsenic (34.6 $mg \cdot kg^{-1}$ d.m.).), tin (23.8 $mg \cdot kg^{-1}$ d.m.), molybdenum (247 $mg \cdot kg^{-1}$ d.m.), cobalt (31.2 $mg \cdot kg^{-1}$ d.m.), vanadium (6.8 $mg \cdot kg^{-1}$ d.m.), strontium (3.1 $mg \cdot kg^{-1}$ d.m.), and barium (7.1 $mg \cdot kg^{-1}$ d.m.). Manganese and phosphorus were also determined in the sample. The mineral waste contained inorganic fluorine in the amount of 210 $mg \cdot kg^{-1}$ d.m., sulphites (98,000% of dry mass) and sulphates (8.6% of dry mass) and a smaller amount of total organic carbon (3% dry mass). The content of heavy metals in mineral waste, at this stage of testing, requires the operator of the cogeneration installation to apply a procedure for dealing with hazardous waste. This is to ensure that special care is taken in the management of this waste in order to prevent its release into water and soil. The threat from heavy metals results directly from their movement in the soil–plant–animal–human trophic chain and from the possibility of accumulation in the last link, i.e. the human body (Dal'nova et al. 2018; Ociepka-Kubicka and Ociepka 2012).

Results of the solid waste fraction test—the third series of research

The most popular method of waste disposal in Poland is still landfilling of waste other than hazardous or inert (Regulation 2015b). Such landfills produce landfill leachate, gases and other forms of pollutants, such as the waste presented in the paper, produced in the heat exchanger of the CHP unit. In the research, the authors attempted to classify the above-mentioned waste by testing extractable metals, non-metallic inorganic parameters and physical parameters (Table 3) according to the Regulation of the Minister of Economy of January 16, 2015 on the types of waste that can be disposed of in a landfill in a non-selective manner (Regulation, 2015a).

The mineral waste settling in the exhaust manifold of the heat exchanger subjected to tests showed high content of heavy metals, including: antimony (220 mg•kg-1 DM), arsenic (75 mg•kg-1 d.w,), chromium (3300 mg•kg-1 DM) 1 d.m.), zinc (316 mg•kg-1 d.m.), nickel (3740 mg•kg-1 d.w.) and other harmful compounds, as in the case of testing waste sample No. 2 (Table 2). Numerous studies on the effects caused by heavy metals prove that, depending on the type, concentration in the environment and the forms in which they occur, heavy metals may have a toxic effect on living organisms, being very harmful to humans and animals, but less harmful to growth and development. plants (Wang et al. 2003; Backer et al. 2006).

In order to identify the type of waste, the provisions of the Act on waste were applied, which define when the waste becomes hazardous waste (Act on waste 2012). The conducted analysis showed that we are faced with hazardous waste generated in the exhaust manifold of the heat exchanger of the cogeneration unit that uses landfill gas to generate energy. Based on the waste catalogue, this waste was classified as: 10 01 18 * wastes from gas cleaning containing hazardous substances (Regulation 2020).

When comparing the test results of the waste samples obtained from the heat exchanger collected after 30,000 operating hours (series 2, sample 2) and after 45,000 operating hours (sample 3, scope 1), an important dependence (especially for CHP unit operators) was observed, i.e. an almost twofold increase in the concentrations of the tested metal parameters and non-metallic inorganic parameters in the sample No. 3, collected after 45 thousand operating

Table 3 Comparison of test results for waste code 10 01 18* with the criteria for accepting hazardous waste for storage at the hazardous waste landfill in the third series, sample no. 3, scope 2

Parameter	Unit	Result	Permissible leaching limit values
Parameters basic			
As	mg•kg^{-1} dry weight	35	25
Ba	mg•kg^{-1} dry weight	4.81	300
Cd	mg•kg^{-1} dry weight	0.926	5
Cr total	mg•kg^{-1} dry weight	3200	70
Cu	mg•kg^{-1} dry weight	132	100
Hg	mg•kg^{-1} dry weight	0.0015	2
Mo	mg•kg^{-1} dry weight	355	30
Ni	mg•kg^{-1} dry weight	3640	40
Pb	mg•kg^{-1} dry weight	3.22	50
Sb	mg•kg^{-1} dry weight	8.54	5
Se	mg•kg^{-1} dry weight	0.499	7
Zn	mg•kg^{-1} dry weight	392	200
Chloride	mg•kg^{-1} dry weight	239	25,000
Fluoride	mg•kg^{-1} dry weight	633	500
Sulphate (SO_3^{2-})	mg•kg^{-1} dry weight	231,000	50,000
DOC	mg•kg^{-1} dry weight	5690	1000
TDS	mg•kg^{-1} dry weight	301,000	100,000
Parameters additional			
LOI	% dry weight	33.2	10%
TOC	% dry weight	6.3	6%
ANC (acid neutralization capacity)	mmol•dm^{-3}	0.149	To obtain the pH 7

hours in relation to the sample number 2, collected after 30 thousand operating hours. This condition is a consequence of the fact that in the period between the second and third sampling, reduced efficiency of LFG purification by the activated carbon bed was found. This situation also results from the fact that new degassing wells have been put into operation at the landfill, and the gas obtained from them is characterized by an increased content of all gas parameters. Additionally, fluctuations in the methane content in LFG had a decisive impact on the energy value of the fuel mixture, which could affect the combustion process in the gas engine, including the resulting fluctuations in the exhaust gas temperature.

The hazardous waste, defined in effect of the research, with the code 10 01 18*, generated during the production of renewable energy in a cogeneration unit, must be managed in a manner that is safe for people and the environment, in accordance with the waste conduct hierarchy. Heavy metals found in hazardous waste are not subject to biological degradation. Their detoxification by organisms involves "hiding" active metal ions within proteins, such as metallothioneins, or depositing them in an insoluble form in intercellular granules for long-term storage or excretion (Ware 2006; Ociepka-Kubicka and Ociepka 2012). One of the ways of neutralizing hazardous waste is the possibility of its storage in hazardous waste landfills, provided that specific requirements regarding the acceptance of waste for storage are met (Regulation Commission 2015).

Leaching test results

One of the elements of waste characteristics, defined by law (Regulation, 2015b), is the leachability of metals, cations and anions containing oxygen as well as dissolved and solid organic compounds. This assessment is used to determine the foreseeable changes in the chemical composition of the leachate generated during landfilling or while using any type of waste. Therefore, in order to check whether the waste from the heat exchanger can be stored in a hazardous waste landfill, basic and additional parameters were tested for the basic leaching test for granular waste having small dimensions of individual elements, with the liquid to solid ratio of 10 dm^3 per kg, expressed in mg•kg^{-1}d.s. The obtained test results were compared with the permissible limit values of leaching for the basic test, and it is presented in (Table 3) in order to allow waste to be deposited in landfills of a given type (Council Decision 2002).

The leaching process in a landfill involves the transfer of dissolved components from the waste to the liquid by percolation or diffusion, e.g. in the event of contact with surface, rain or underground water (Szymański and Janowska 2016). The solubility of waste components in liquid depends on the environmental conditions, chemical factors, i.e. the content of other accompanying chemical compounds in the waste and contact liquid, as well as on physical and biological properties. Factors that strongly affect the solubility of inorganic compounds include the pH and redox potential, the phenomenon of adsorption, the formation of complex compounds and the common ion effect, as in the case of arsenic, which, when adsorbed on iron oxides, is released from oxidized deposits during the transformation between oxidizing and reducing conditions. And the solubility of organic components is influenced by the polarity and the partitioning effect (Makowska et al. 2018; US Report 2003). The obtained test results showed exceedance in the case of 11 basic parameters (As, Cr, Cu, Mo, Ni, Sb, Zn, Fluoride, DOC, TDS, sulphates) for the leaching test above the limit values. Figure 4 presents the values of 8 parameters obtained in effect of the test, with the comparison to permissible values.

Hazardous waste from a heat exchanger is characterized by an increased content of ecotoxic elements, i.e. heavy metals (arsenic, chromium, copper, nickel, zinc), molybdenum, antimony, as well as fluorides, sulphates, dissolved organic carbon and solid dissolved compounds. The lowest allowable arsenic content in hazardous waste should not exceed 25 mg•kg^{-1} dry weight (Regulation 2015b), and in the case of the tested wastes, this value was 35 mg•kg^{-1} dry weight. Natural sources of arsenic include: volcanic eruptions, sea surface waves, microbiological processes and others.

The anthropogenic sources of arsenic include: coal combustion, mining of mineral resources, production of batteries, soil fertilization, steel and metallurgical industries. Arsenic, apart from mercury, cadmium and lead, is one of the most toxic elements for the environment. This element is characterized by high mobility in the water–soil environment, which is essential in terms of its storage (Parth et al. 2011; Loeppert et al. 1999). The content of antimony in the tested waste was 8.54 mg•kg^{-1} dry weight, with the permissible value being 5 mg•kg^{-1} dry weight. Antimony is introduced into the atmosphere with exhaust gases from metallurgical plants and furnaces for fuel materials and garbage. Its origin is believed to be in the stored waste from the aforementioned industries. Scientists have also demonstrated an increase in the concentration of antimony in heavy soils and those rich in organic matter (Liu et al. 2022; Niedzielski et al. 2000). Atmospheric air in urban agglomerations may contain several times higher concentrations of antimony than the concentrations of other trace elements. Antimony may enter groundwater together with organic humic acids (Szymański 2009). Figure 5 presents the values of the other 3 parameters showing exceedance found in effect of the tests in terms of permissible values.

Waste stored in landfills may take a heterogeneous multiphase form, i.e. it may be mixed with soil material (organic and inorganic) of various origins, and it may undergo various changes in its qualitative composition under the influence of the aforementioned physical and chemical factors as well as due to the impact of microorganisms and oxygen content. The tested waste showed an excessively high content of dissolved organic carbon (DOC) in the amount of 5,690 mg•kg^{-1} dry weight (permissible value is 1000 mg•kg^{-1} dry weight), high content of total dissolved solids (TDS) in the amount of 301,000 mg•kg^{-1} dry weight, with an acceptable leaching limit of 100,000 mg•kg^{-1} dry weight, and an increased content of sulphates (SO_3) 231,000 mg•kg^{-1} dry weight and fluorides 633 mg•kg^{-1} dry weight. Increased values of sulphate indicate that materials from the metallurgical industry, e.g. metal colouring, are stored in the landfill. The group with raised tested concentrations includes also heavy metals, i.e. total chromium, the concentration of which is 3200 mg•kg^{-1} dry weight, copper (132 mg•kg^{-1} dry weight), zinc (392 mg•kg^{-1} dry weight) and nickel (3640 mg•kg^{-1} dry weight), which can form organometallic and mineral bonds (Jianguo et al. 2004).

The combination of sulphuric acid and copper, i.e. copper sulphate, present as a mineral called chalcanthite, is effectively used in the metallurgical industry, e.g. for colouring iron. It is also used on a large scale in the production of paints and varnishes as well as for wood preservation in the wood and furniture industries. In addition, copper compounds are used in the pharmaceutical, horticultural and agricultural industries due to their bactericidal properties.

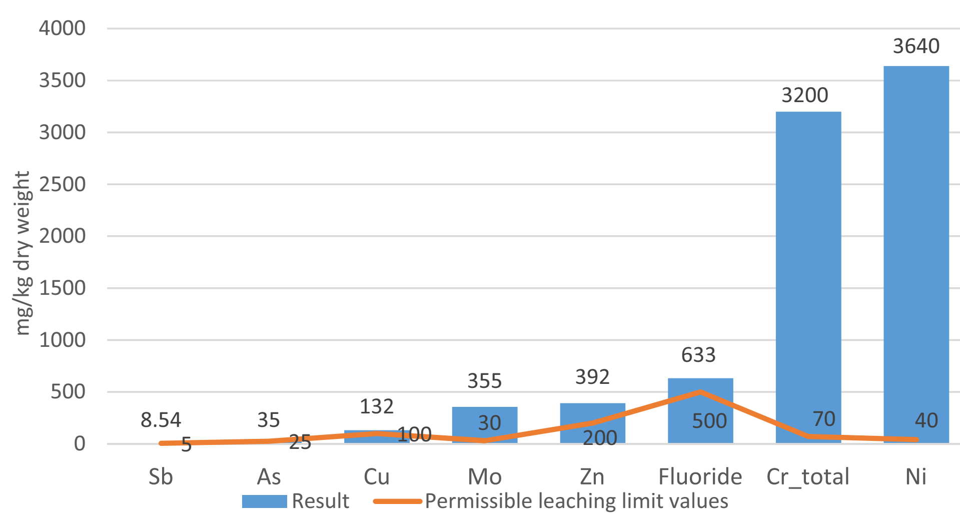


Fig. 4 Exceedance of eight basic parameters for the leaching test

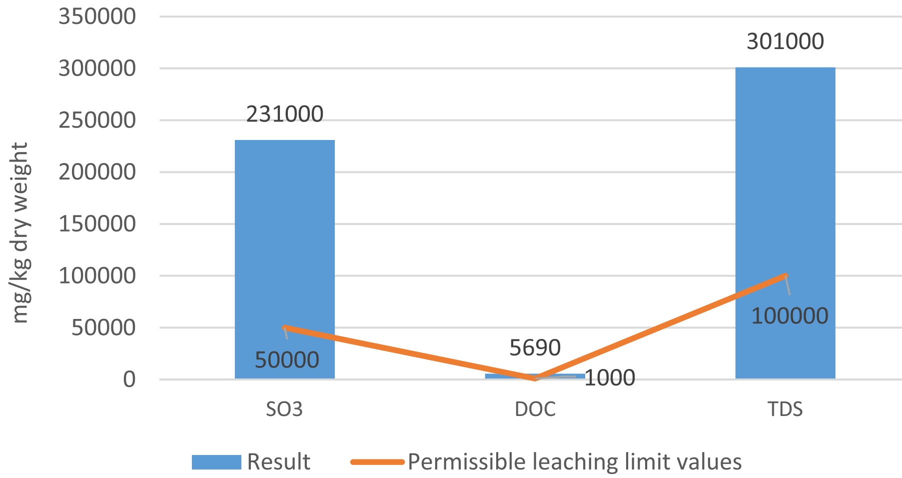


Fig. 5 Exceedance of 3 basic parameters for the leaching test

Similarly, zinc sulphate is successfully used, for example, as an electrolyte component in electrogalvanizing. Heavy metals in the aquatic environment usually form dissolved metal forms, while in the ground, they are poorly soluble. In municipal landfills, heavy metals usually occur in both soluble and poorly soluble form, which is also confirmed by the results of the waste tested for the content of DOC and TDS (Jianguo et al. 2004; Mu et al. 2018).

Waste neutralization methods

The test results for waste with the code 10 01 18 *, being a mineral deposit, taken from the heat exchanger installed in the exhaust manifold of the gas engine of the cogeneration unit, do not meet the requirements for storage in a hazardous waste landfill. Therefore, other methods of its neutralization should be sought, e.g. further treatment or storage in an underground storage facility for hazardous waste. Figure 6 shows the procedure to apply when dealing with hazardous waste when it is stored in hazardous waste landfills of a given type (Council Decision 2002).

Hazardous waste may be stored in an underground hazardous waste landfill, provided that certain conditions are met. The criteria to be met to allow for the dispatch of waste for storage in an underground hazardous waste landfill comprise: permissible leaching limits and additional parameters. Acceptable limit values are considered to be met when the values are three times higher than those specified in Table 4 as the acceptable values for the leaching limit (Regulation 2011). Despite this rule, three times higher than the acceptable parameters of the leaching test results showed that 6 parameters of the waste were exceeded, i.e. total chromium, molybdenum, nickel, sulphates, dissolved organic carbon (DOC) and total dissolved solids (TDS). This fact does not exclude the possibility of storing waste with the code 10 01 18 * in an underground hazardous waste dump. In a situation where the following conditions are jointly met: there is a natural geological barrier, the conditions specified in the hydrogeological and geological-engineering documentation are met and the waste has been subjected to the procedure of admitting waste to the underground landfill, storage is allowed (Grygorczuk-Petersons and Wiater 2016).

Discussion

A cogeneration unit powered by landfill gas, producing renewable energy, is also a place where waste is generated, including hazardous waste, which should be identified and examined in order to determine the proceedings to follow in terms of their disposal. Identified waste in quantitative and qualitative terms should be included in the overall balance of waste generated in the landfill and indicated in relevant reports and documentation, including fees for using the environment. Cogeneration installations operated in landfills are a source of green energy generation, but they also have a negative impact on the environment. This applies mainly to waste, including hazardous one, generated during the production of electricity and heat from landfill gas, which is in fact a source of renewable energy. The article fills the gap in terms of the identification, research and neutralization method of hazardous waste, which is a mineral deposit formed in the heat exchanger on exhaust gases in the exhaust manifold of the CHP unit. Due to the fact that cogeneration installations powered by landfill gas are not equipped with exhaust gas reduction systems, gaseous and dust pollutants settle in the heat exchanger by gravity. The exchanger is a structural obstacle in the exhaust manifold, accumulating mineral deposits, which consequently reduce the heat exchange in the system exhaust gas—heating medium. To prevent such problems, systematic cleaning of the heat exchanger in question is required by removing mineral deposits mechanically or hydraulically.

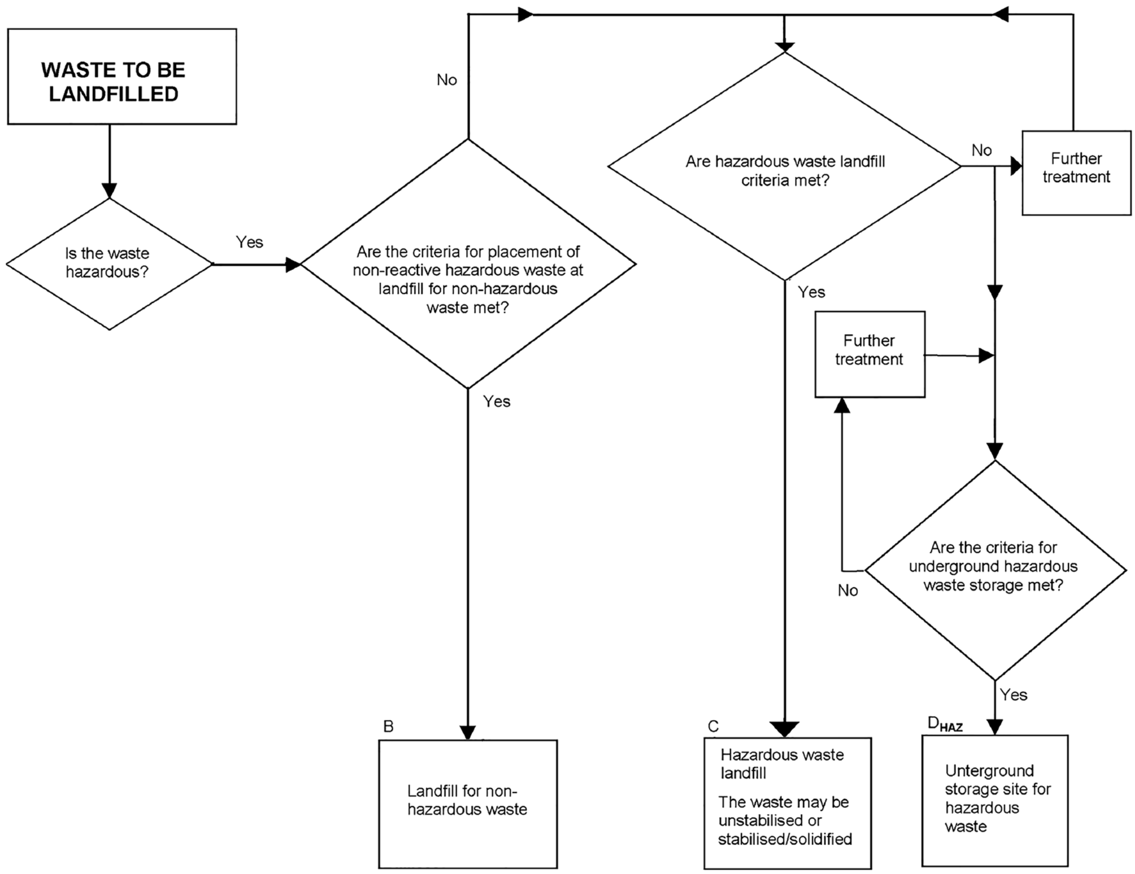


Fig. 6 Procedure for dealing with hazardous waste in order to store it in hazardous waste landfills

In the literature on the subject, no publications have been found directly indicating this type of action improving the efficiency of heat generation in the CHP unit powered by landfill gas. And a more important problem for operators of CHP installations powered by landfill gas is the lack of detailed studies of such waste in terms of its neutralization as hazardous waste. The analysed publications focus only on general studies of the composition of mineral deposits, on comparing them depending on the type of biogas, without indicating the methods of their neutralization, safe for the environment and humans. In the work, Konkol et al. (2022), the authors performed laboratory tests of mineral deposits collected, among others, from the exhaust manifold of a gas engine powered by landfill gas. The test results showed the presence of the compounds of sulphur (0.02–0.84%), arsenic (0.4–1.1%), silicon (81%), antimony (17%) and iron (0.34%) in the deposits. The authors suggest that in order to limit the development of mineral deposits in the engine and in the exhaust manifold, landfill gas should be treated in several stages, and engine oil enhancing additives specially selected for the type of gas burnt should be used, as recommended by the manufacturer for a given type of engine.

The authors decided to reveal the lack of such research and describe this fact on the example of own research of a real CHP facility, taking into account scientific reliability and the need to tighten the waste balancing and reporting system. This fact applies not only to CHP units powered by landfill gas, but also to other types of biogas, including those from sewage treatment plants, municipal, agricultural, food industry and other biogas. Operators of CHP installations powered by biogas include hazardous waste in their reports, such as used activated carbon, used engine oil, and oil filters, while hazardous waste generated in the exhaust manifold and heat exchanger system is omitted.

The problem of testing the composition and potential utilization methods of hazardous waste, namely ashes from hazardous waste incineration, was examined, among others, by Liu H. et al. (2022). The results of the analyses of waste burnt in a rotary kiln with a capacity of 100 Mg per day showed that the highest percentage share in the ash from bag filters was reported for Na, S and Cl (over 70%). And Cu, Mn, Cr were deposited in greater amounts in the bottom ash due to their low volatility, while Zn, Pb, Cd, Se and As were more often present in ash particles from bag filters. However, it was found that chemical adsorption in

Table 4 Test results of exhaust gas cleaning waste in the exhaust manifold in the third series, sample No. 3, scope 1

Parameter	Unit	Result	Measurement uncertainty
Extractable metals/mainly cations			
Sb	mg•kg^{-1} dry weight	220	±43.9
As	mg•kg^{-1} dry weight	75.0	±15.0
Ba	mg•kg^{-1} dry weight	15.5	±3.09
Be	mg•kg^{-1} dry weight	<0.010	–
Cr total	mg•kg^{-1} dry weight	3300	±659
Sn	mg•kg^{-1} dry weight	52.8	±10.5
Zn	mg•kg^{-1} dry weight	316	±63.2
P	mg•kg^{-1} dry weight	35.9	±7.2
Cd	mg•kg^{-1} dry weight	<0.40	–
Co	mg•kg^{-1} dry weight	64.8	±13.0
Li	mg•kg^{-1} dry weight	<1.0	–
Mn	mg•kg^{-1} dry weight	2110	±423
Cu	mg•kg^{-1} dry weight	148	±29.6
Mo	mg•kg^{-1} dry weight	478	±95.7
Ni	mg•kg^{-1} dry weight	3740	±747
Pb	mg•kg^{-1} dry weight	3.7	±0.7
Hg	mg•kg^{-1} dry weight	<0.20	–
Ag	mg•kg^{-1} dry weight	<0.50	–
Sr	mg•kg^{-1} dry weight	7.1	1.42
Tl	mg•kg^{-1} dry weight	<0.50	–
V	mg•kg^{-1} dry weight	14.7	±2.94
Fe	mg•kg^{-1} dry weight	53,600	±10,700
Non-metallic inorganic parameters			
Chlorides	mg•kg^{-1} dry weight	<40	–
Total inorganic fluorine	mg•kg^{-1} dry weight	460	±140
TOC	% dry weight	6.3	±0.94
Sulphates (SO_4)	% dry weight	20.2	±2.02
Sulphates (SO_3)	% dry weight	231,000	±34,600
Physical parameters			
Bulk density	g•dm^3	460	±70
LOI at 550 °C	% dry weight	33.2	±1.66
Dry weight at 105 °C	%	97	±5.85

the range of medium and high temperatures was the cause of increased concentrations of As in the ash from the boiler. The content of heavy metals, including e.g. Pb, Cd, Zn, Cu, Sn and sulphates in fly ash in electrostatic precipitators from municipal waste incineration has been reported by several research teams. Scientists also proposed a new approach to how to stabilize metals present in the ashes to minimize the leaching process. They synthesized a new type of chelating agent for heavy metals that proved to be more effective than standard inorganic chemicals such as sodium and calcium sulphides. In addition, the waste stabilized with chelating agents met the standards for the control of waste storage in terms of the presence of heavy metals. A pH-dependent leaching experiment showed that stabilized fly ash with the use of a heavy metal chelator maintained long-term stabilization over a wide range of pH values. Thus, the risk of secondary contamination of stabilized products was reduced in the event of a change in environmental conditions during the period of its disposal (Jianguo et al. 2004; Wolffers et al. 2021). Wang et al. (2022) investigated the factors influencing the content of metals in soil in municipal landfills. The test results showed that Cr and Zn are the main soil contaminants. Differences were found in the content of chromium Cr, Hg, Pb, Zn and As between soil samples from sanitary (SL) and non-sanitary (NSL) landfills. The concentration of As in soil showed a significant positive correlation ($r = 0.472$) with rainfall in NSL.

Contemporary scientists are widely discussing the topic of high-efficiency heat exchangers used for waste heat recovery and their energy optimization (Zhang et al. 2022; Shi et al. 2022; Jiang et al. 2022; Chen et al. 2022). And the literature lacks qualitative research on waste from heat exchanger deposits in cogeneration systems powered by landfill gas. Research of this type has not been performed on a large scale. It is also difficult to find information on leaching tests for such waste for storage or disposal purposes, when the waste does not meet quality requirements for a hazardous waste landfill, and therefore the research carried out by the authors is pioneering in this respect.

When focusing on generating electricity and heat from landfill gas, which is a source of renewable energy, CHP plant operators should not forget about waste generated in such a process and the need to examine, record and properly dispose of it. Therefore, the operator of the landfill in which the cogeneration unit is operated will be forced to review the current amount and type of waste generated as part of the operation of the facility, which is a waste landfill. Depending on the provisions of the servicing agreement for the cogeneration unit, it is also possible to take over the waste management obligations from the heat exchanger by the entity performing inspections and repairs on the basis of the waste transfer note.

One should not ignore the fact that the production of hazardous waste in CHP powered by landfill gas in the amount of 0.005 g/kWh is a marginal value. The key issue in this matter is the economy of scale. In the published work (Ciuła et al. 2023a, b), it was shown that in 2021, 1,345.58 GWh of eclectic energy from all types of biogas was produced in Poland. This fact shows that in Poland in 2021, 6,727.8 kg of hazardous waste was generated in CHP installations, about which there is no information in official reports. The scale effect becomes particularly important when we start to balance the electricity generated on a European and global scale and the related amount of hazardous waste generated in heat exchangers.

The results of the research on the deposit from heat exchangers, including the leaching test, unequivocally showed that waste with the code 10 01 18 * is a hazardous waste that cannot be stored in a hazardous waste landfill, as it contains significant exceedances of the permissible parameters. Therefore, other methods of neutralizing such a waste should be sought, including e.g. storage in underground hazardous waste dumps or its further treatment.

Summary

The acquisition and use of landfill gas as a source of renewable energy for energy purposes in a cogeneration system at a landfill site are an optimal solution in this regard. However, it should be borne in mind that in this positive process of green energy production, hazardous waste is generated, which should be identified and disposed of under appropriate conditions. Pioneering research of waste being a mineral deposit, collected from the heat exchanger installed in the exhaust manifold of a gas engine of the cogeneration unit, showed high concentrations of ecotoxic elements, i.e. heavy metals (arsenic, chromium, copper, nickel, zinc), molybdenum, antimony, having carcinogenic effects. High concentrations were also reported for fluorides, sulphates, dissolved organic carbon and dissolved solids. Considering the above, the waste is classified as 10 01 18 *: wastes from gas cleaning containing hazardous substances. The conducted leaching test, aimed at determining an appropriate method of neutralizing waste with the code 10 01 18 *, showed that the permissible concentration was exceeded for 11 parameters (As, Cr, Cu, Mo, Ni, Sb, Zn, F-, DOC, TDS, and sulphates), which disqualified the waste for storage in a hazardous waste landfill. The analysis of the waste parameters for its storage in the underground storage facility for hazardous waste showed that the permissible concentration for 6 parameters (Cr, Mo, Ni, DOC, TDS, sulphates) was exceeded. However, in this case, storage may be accepted on the condition that the landfill in question has a natural geological barrier that meets the conditions set out in the hydrogeological and geological-engineering documentation, and that the waste has been subject to the procedure of admitting waste to the underground landfill.

The longer the deposition time of the mineral deposit in the heat exchanger, the higher the concentration of heavy metals and other harmful compounds in the waste, and the more difficult it is to manage and/or dispose of such waste. Therefore, the activities involving the cleaning procedure of the CHP unit should be permanently included in the schedule of people operating the WWTP.

Hazardous waste generated in the municipal economy and the energy industry using landfill gas to generate renewable energy pose a threat to humans and the environment. Regardless of the amount of waste generated in a given process, actions are necessary to limit their negative impact. Hazardous waste generated in the heat exchanger of the exhaust manifold of a gas engine of a cogeneration unit is characterized by an increased content of ecotoxic elements, i.e. heavy metals (arsenic, chromium, copper, nickel), molybdenum, antimony, as well as fluorides, sulphates, dissolved organic carbon and solid compounds. dissolved. Failure to correctly identify this waste during operation and renovation work of a cogeneration unit powered by landfill gas may consequently lead to the selection of an inappropriate form of waste disposal. In such a case, their release into the soil, surface or groundwater may lead directly to contamination of the natural environment. An indirect effect of such an event may be the entry of heavy metals into the bodies of people and animals, most often through the food route. The health effects of regularly consuming products containing even small amounts of these elements may become apparent after many years. Therefore, it is important in this process to develop appropriate procedures for dealing with particularly hazardous waste, such as sludge from a heat exchanger. A key element in the generation of hazardous waste is compliance with the deadlines for inspections and renovations of cogeneration units, along with maintaining appropriate technical documentation and balancing of this waste.

The identified waste in quantitative and qualitative terms should be included in the overall balance of waste generated in the cogeneration unit at the landfill and indicated in relevant documentation and reports. The research and analyses performed in this work will serve as input material for defining the methods of hazardous waste disposal from the CHP unit.

Author contributions CRediT authorship contribution statementJózef Ciuła contributed to conceptualization, methodology, investigation, formal analysis, data curation, and writing—original draft preparation. Agnieszka Generowicz contributed to conceptualization, methodology, investigation, formal analysis, writing—original draft preparation, and supervision.Iwona Wiewiórska: conceptualization, methodology, investigation, formal analysis, data curation, and writing—original draft preparation.Krzysztof Gaska contributed to validation, software, investigation, resources, visualization, and project administration.Anna Gronba-Chyła contributed to validation, investigation, resources, visualization, and software.Monika Golonka contributed to validation, investigation, data curation, and project administration.Agnieszka Makara contributed to formal analysis and data curation.All authors have read and agreed to the published version of the manuscript.

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Declarations

Conflict 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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