Exploatarea potențialului energetic al gazelor de la depozitul de deșeuri din materiale organice derivate din deșeurile municipale - folosind combustibil alternativ durabil în instalațiile de utilizare a gazelor naturale

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Abstract. In the light of the increasing exploitation of natural resources, a rigorous approach and an effective identification of practical solutions needed to ensure the conservation of the environment is required, given the crucial importance of this issue. This scientific work explores an innovative method of treatment and improvement of landfill gas produced from municipal and similar wastes, for a carefully selected and thoroughly studied landfill, in order to obtain a sustainable alternative fuel that can be used in natural gas installations. The method analyzed in this study is called high-pressure water scrubbing, which allow the production of biomethane. Additionally, using the mathematical model LandGEM, the energy potential and gas generation rate was determined, for a selected landfill located in Alba Iulia, Romania country. This article also provides a review of technology that uses the high-pressure water scrubber for the integration of improved landfill gas into the mobile cylinder. Through this investigation, the landfill in Alba Iulia, Alba administrative territorial unit, Romania, can be considered as an energetic resourse who can be used through a sustainable potential solution for protecting current natural resources.

Keywords: organic material, sustainability, natural gas, fuels, utilization installation.

Nomenclature	
Adsorption column - AC	
Atmospheric pressure - AP	Methane - CH ₄
Carbon dioxide - CO ₂	Residual gases - RG
Compressed gases - CG	Regenerative adsorption - RA

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Compressed biomethane storage reservoir –	
CBMSR	
Desorption column - DC	
Flash Column - FC	Storage reservoir - SR
High pressure - HP	Single-pass water scrubber - SPWS
High-pressure water scrubber - HPWS	Water scrubber - WS
Hydrogen sulphide - H ₂ S	
Landfill gases - LFG	

Introduction

European Union waste landfill practices [1] emphasize the importance of preserving the environment, protecting the health of participants and using natural resources rationally.

The sustainability of the gas sector is currently the subject of intensive debates, due to the supplier dependency and the increasment of the prices market. In Romania, current energy strategies focus on the use of fossil fuels as a raw material for heat production [2]. However, innovative national technologies that would allow energy from waste are still not very applicable and need further development. Therefore, a careful assessment on options available to reduce the environmental impact of the energy sector and encourage sustainable development is needed.

Currently [3] there are 47 compliant municipal landfills in Romania at the level of year 2022. In the reference period 2006-2018, 56 non-compliant landfills were registered and had to stop their activity, being permanently closed. Due to the fact that landfills that stopped their activities were not permanently monitored, there are no statistics available at the national level on the amount of total existing municipal waste (t/year).

In 2020, according to statistics [4] of municipal waste deposited in operation landfills, an amount of about 286 kg/capita of waste was recorded [5]. For a population of about 19.02 million inhabitants [6], in December 2021 Romania is estimated to have produced about 19,290 t/year of waste, an upward trend compared to the period 2015-2019, when about 259 kg/capita of municipal waste was recorded [7].

At the same time, the target to be achieved by 2025 [8] is to reduce the amount of municipal waste left in landfills by up to 55% by mass for energy recovery or recycling. In the same order, the amount of municipal waste landfill should be reduced by 60% until 2030 and 65% until 2035. At national level, we have no information on the potential for obtaining alternative fuels from municipal waste for collection and storage in SR and mobile cylinders.

Case study: Investigating the current state in Alba Iulia landfill

This scientific article provides an investigation of the current situation of a noncompliant landfill site, closed permanently in 2015 in Alba Iulia, Alba County, Romania. In addition, a possible technical solution has been identified to use the landfill gas as an alternative fuel, compared to the traditional solution based on natural gas from fossil fuels.

Using the LandGEM calculation software, it has made an estimate of gas emissions from municipal solid waste to evaluate the energy potential with a reference period of at least 100 years from the opening of the landfill.

The area occupied by waste of the non-compliant landfill in Alba Iulia that stopped its activity was, before the final closure in 2015, 5,99 ha, and after the systematization by compacting the waste mass, the surface of the site became 3,47 ha. The landfill capacity is 660 000 cubic metres and the amount of waste landfilled is approximately 238 260 t. The types of waste are municipal and similar waste, classified in the "b" category of importance – non-hazardous waste landfills. Waste disposal has not been carried out taking into account selective collection. The period of storage of these wastes in the mentioned location was more than 30 years.

In the Figure 1 could be observed the identification of the area studied, who is composed by four parts: the geographical location, the territorial delimitation where the landfill is located, its location and a general view taken on site.

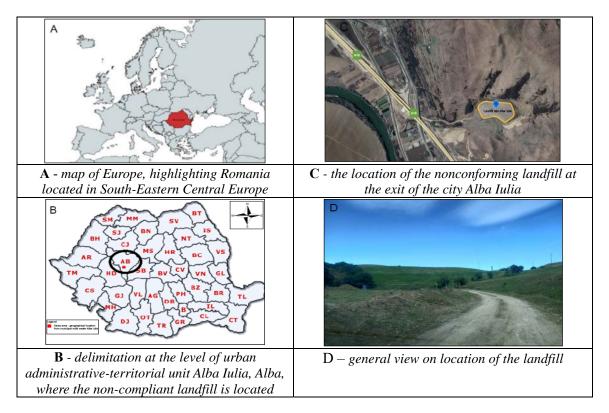


Figure 1 - Location of the study area

Upon completion of the landfill closure process, the landfill gas collection and combustion system was implemented, consisting of 14 landfill gas collection and transfer wells, 1,370 m collection pipelines, landfill gas collection station and gas combustion unit with a capacity of 150 Nmc/h.

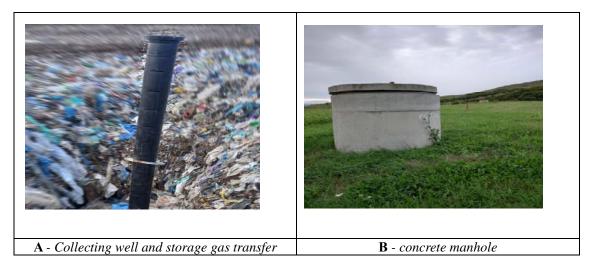
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The 14 storage gas collection and transfer wells have the role of collecting LFG inside the waste mass, with LFG transmission system going to the unit with a flame for combustion. LFG collection and transfer wells are made up of boreholes drilled directly into the waste mass.

The pipeline networks are of the branched type, connected to the LFG gathering and transfer wells, these are also connected to the storage gas gathering and transfer station.

The LFG collection and transfer lines are connected to a manifold, and from this main manifold they are distributed to the combustion unit via a pipeline. The LFG flame combustion unit is provided for the purpose of controlled combustion of surplus gas and is equipped with a blower and combustion control system.

Below, in Figure 2 and Figure 3 is shown the components of the landfill gas collection and transfer system site which include: the LFG collection and transfer well, the concrete manhole, the combustion unit and the gas collection station.



 $Figure \ 2- Collecting \ well \ and \ storage \ gas \ transfer, \ concrete \ manhole$



Figure 3 - Landfill gas combustion unit and landfill gas collection station

The main component of landfill gas is CH₄, followed by CO₂. The other component gases are trace amounts of hydrogen sulphide (H₂S), oxygen (O₂), and the

rest of the gas mixture is of unknown composition. In the raw phase, the CH_4 concentration is approximately 40-70% and the CO_2 concentration approximately 30-60% [9]. The other secondary constituents are expressed in ppm.

According to the 2015-2022 Test Reports [10], regarding the generation rate of CH₄ in landfill site in Alba Iulia, it reached an average value of 65%. The composition of CH₄ falls within the range of 40-70%, reaching phase IV – the stable methanogenic phase.

Among the key factors that favoured the production of LFG within the noncompliant landfill waste mass are:

- bacteria content - methanogenic bacteria that favoured the methane formation process;

- nutrient content - present in organic waste;

- temperature - anaerobic bacteria have an optimal production range of 30-41°C while their activity decreases at temperatures below 10°C;

- waste category - municipal, non-hazardous;

- waste age - uncertain, over 30 years;

- landfill classification - the landfill is a standard landfill with a high initial moisture content, and no separate collection of household waste was ordered before the shutdown [9].

Energy potential and landfill gas generation rate

To estimate the production of landfill gas from Alba Iulia landfill, the LandGEM mathematical model developed by US EPA it was used. This model assumes a first-order equation quantifying emissions from municipal solid waste landfills, called DMS. This mathematical model is preferable when information on landfills is limited. The calculation formula used is:

$$Q_{CH_4} = \sum_{i=1}^{n} \sum_{j=0.1}^{1} k \cdot L_0 \cdot \left(\frac{M_i}{10}\right) \cdot e^{-k \cdot t_{ij}} [11, 12], \tag{1}$$

where:

Q_{CH4}- approximate annual methane production in Mg / year;

i - the time period of one year during which the LFG increased;

n - the calculated reference year when the waste is accepted;

j - growth for 0.1 years;

k - constant methane generation rate (an⁻¹);

Lo - potential methane generation capacity in m³/Mg;

M_i -mass of waste accepted in year i, expressed in Mg;

t_{ij} - age of section j of the waste mass M_i, accepted in year i.

The LandGEM mathematical model automatically calculates landfill gas emissions, taking into account the annual removal rate, time variation and total landfill capacity. The main parameters used are "Lo" - potential CH₄ generation capacity of the waste, and "k" - CH₄ generation rate over time. The last parameter is the practical reduction of the percentage of landfill gas generation considering its main constituents Diana-Laura Mertan, Adrian Retezan, Daniel Dan, Roxana-Iuliana Blezniuc, Florin Bumbar

when the maximum rate is reached. The simulation LFG production rate in Mg/year is shown in Figure 4.

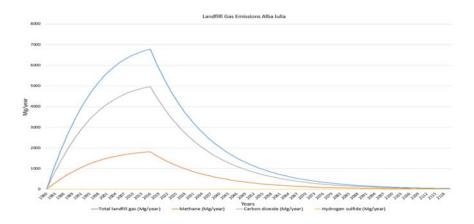


Figure 4 - LFG production rate from Alba Iulia deposit, calculated over a reference period of at least 100 years from the opening of the deposit

The landfill was established in 1980. The waste generation rate was calculated taking into account Eurostat reports [13] on municipal waste generated per person in Romania. It also took into account the nationally declared population census [14] between 1980 – 2015; last year marked the closure of landfill. The maximum LFG production was expected to be completed in 2017, 2 years after the closure of the landfill site, recording about 6 800 Mg, i.e.7 456 t. subsequently, these generation rates of the main constituents from LFG will decrease to about 150 Mg, i.e. 165 t in 2055, as the organic fraction is depleted, decreasing continuously for up to 100 years. At present, LFG effects persist until the inert phase is reached.

Enhancing LFG through water scrubbing system: A Pathway to Biomethane Production

In order to improve the quality of raw LFG, the treatment process is indispensable to remove contaminants and moisture content. This operation is necessary for the treated LFG to obtain biomethane.

In order to obtain purified LFG - biomethane, a technological process must be adopted from the LFG treatment technologies. The chosen method, called water scrubbing, is based on the physical effect of dissolving gases in the liquids with which they come into contact. This type of installation required in Alba-Iulia is designed and shown in Figure 5.

Following the application of the LFG treatment and improvement method by WS system [15, 16], located in Alba-Iulia deposit, the concentration of CO₂ and other contaminants can be reduced to less than 2%, obtaining a methane purity at least 97%.

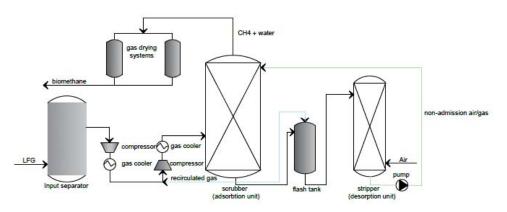


Figure 5 – Design of high-pressure water scrubber installation for Alba Iulia landfill

This method was chosen as a result of comparing several methods involving CH₄ treatment and enhancement, and is a simple and less expensive method than the other methods, such as physical adsorption, chemical adsorption, pressure swing adsorption, membrane separation, and cryogenic separation [17].

The chosen process also includes several ways in which the operation can be achieved: the water used to remove unwanted chemicals can be used once (SPWS) or the already used water can go through a regenerative process (RA) [9, 18]. The first option implies that the water requirement will be higher than regenerative process, but in this case, there will be no possible contamination of the water with traces of H₂S and CO₂. This method is adopted when H₂S concentrations are low. Regenerative absorption means that the water requirement used in the methane treatment and upgrading process will be much lower [19].

The WS can also be performed at AP or HP (6-10 bar) [20, 21, 9], with operating pressures and temperatures provided by each equipment manufacturer. The compression of raw gases is carried out in two stages, using the two connected compressors connected in series, up to 4-8 bar and partly up to 10 bars [22]. After each compression operation, cooling is achieved and then enters the AC. The technique using HPWS has the advantage that LFG enters AC at HP, thus increasing the dissolution rate of the gas in the water it comes into contact with, implying a smaller amount of liquid required [23]. Due to the fact that CH₄ is only partially soluble in water, the saturated water is passed to the FC, which operates in the pressure range 2-4 bar in order to separate the dissolved CH₄, and is then directed to the AC [22]. After these operations, the saturated water passes to the DC, making contact with air for the water desorption process. It should be noted that the efficiency of the desorption process is recorded when the temperature is low [20].

This process does not require a preliminary desulfurization step, because H_2S is removed by the AC, and it is also a physisorption process, which involves reversible absorption based on physical binding forces (van der Waals) [22]. However, as mentioned in the following lines, in some situations desulphurization is recommended when it involves recovery of methane from the waste gas stream. Diana-Laura Merțan, Adrian Retezan, Daniel Dan, Roxana-Iuliana Blezniuc, Florin Bumbar

RG dissolved in the mass of the liquid substance are removed toward the ends of the DC. In this process, small concentrations of CH_4 can be removed together with waste gases, and a "catalytic oxidation" is recommended for their treatment, which implicitly requires a previous activity involving desulphurization, since elemental sulphur contamination affects the catalyst [20]. Also, this process of precision desulphurization is recommended to be carried out before starting the actual WS operation, as the manufacturers of this equipment allow a tolerance of 300-2500 ppm H₂S in case of elemental sulfur contamination [15].

Based on *Figure*. 4, the landfill site in Alba Iulia recorded an approximate 39 300 Mg raw LFG, during the reference period of 2017-2022. Of this value, 69% represents CH₄, while 28% represents CO₂. The remaining 3% consists of unwanted constituents. Through the implementation of treatment and improvement method on the raw LFG, approximately 38 121 Mg of usable purified LFG was obtained, representing 97% usage gas.

This emphasizes the superior effectiveness of the realised WS method for purifying the raw LFG, compared to the existing conventional approach of burning the excess gas at the Alba Iulia landfill. The results highlight a significant improvement in LFG purification using WS technique.

Storage in cylinders of LFG resulting from gas treatment and improvement using high pressure water scrubber

After application of the LFG treatment and improvement method by WS at Alba Iulia landfill, which led to improve the composition of the combustible gases generated from organic material in municipal waste, the issue of storage the resulting gases will be further investigated. This is accomplished in two steps using SR and mobile cylinders.

The CBMSR is an ergonomic alternative. From a financial perspective, the compressor is more expensive, as it compresses the resulting gas to the desired pressure, thereby reducing the need for storage. The introduction of improved LFG into SR can be done over short periods of time, in just a few minutes, or over long periods of time, which can reach fullness in a few hours, the long filling cycle having the advantage of lower energy consumption and constant temperature of the introduced gas [24]. At the same time, the SR can be used for high or low pressure applications. Nevertheless, for scenarious involving transportation in locations that necessitate a significant distance, the HP biomethane SR solution is recommended. In the next step, we will connect the HPWS to the stepper gas compressor as shown in *Fig.* 6, together with the HP SR that can operate up to 300 bar, in order to create a resistance to gas flow [24]. For HP storage a cylindrical reservoir was designed. These types of SR vary in terms of their size, shape and storage capacity, alloys used, depending on the equipment put on the market by the manufacturers.

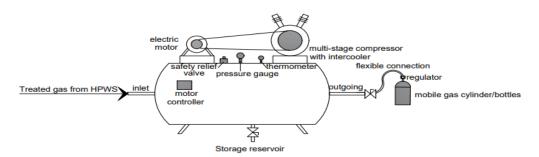


Figure 6 – Design of treated and improved LFG, storage in reservoir and subsequently in mobile cylinder for the Alba Iulia landfill

However, in order for the alternative fuel to reach the beneficiaries, it must be placed in a smaller cylinder to facilitate transport and storage in the buildings. Biomethane bottled in small cylinder provides end-users access to fuel for domestic use and/or heating of buildings, where a transportable cylinder has a pressure greater than 0.5 bar [25]. This type of storage can be obtained after the results of the first stage from the SR, as these reservoirs are used to distribute CG. In this case as well, stepwise compression is required. These cylinders must be equipped with shut-off valves, a gas flow limiting device and a pressure reducing device to correspond to the low working pressures of appliances consuming gaseous fuels for domestic use and/or space heating. The maximum allowable loading pressure is up to 200 bar for CH₄ CG transportable cylinders according to national requirements [26]. As with the compressed biomethane SR, the cylinders differ in size, shape, storage capacity, and alloys used.

Combustible gases obtained from organic materials derived from landfill site, for the purpose of being used by the end consumers, are bottled in small cylinders. The quantity of resulting gases could support a significant proportion of the demand. Knowing that the production of purified LFG obtained trough the HPWS method from the Alba Iulia landfill is approximately 38 121 Mg for the reference period 2017-2022, this would correspond to approximately 1 440 mobile cylinders considering storage purified LFG, each weighing 11 kg.

This has a positive impact on the environment and contributes to supporting sustainability principles. By using gases derived from organic materials, the dependence on fossil fuels is reduced, and the environment is protected. This represents an efficient way to preserve available resources and promote sustainable development.

Conclusions

Landfill gas, as well as the technological system for treating and improving these types of gas, in Romania is not enough discussed. They are seen mostly as generators of hard difficulties and there are no energy recovery solutions for them. Natural gas utilization strategies to improve the energy sector at national level only target the fossil fuels and do not include landfill gas as an alternative fuel. New strategies and policies are needed to regulate the use of auxiliary fuels, i.e. LFG. This requires new methods of treatment-purification, storage and transport of these gases resulting from organic materials to the final consumers for use as alternative fuels for natural gas installations.

Starting from the assumption that CH_4 and CO_2 are the main components of LFG, we showed that using a complex LFG treatment and improvement system, together with the application of the HPWS method, can increase methane purity to 97%, reducing CO_2 concentrations to less than 2%. Using the LandGEM mathematical model, we estimated LFG production, starting from the probable year of its opening and culminating with the moment when it would become inert, while analyzing gas storage solutions, such as cylinders and storage reservoir, in order to ensure access to sustainable alternative energy sources for end-users of natural gas installations.

Analyzing the situation of non-conforming municipal waste landfills in Romania, it is important to take into account that many of them have stopped their activity due to the lack of selective collection at the time of their establishment, the resulting gas still being burned in combustion units and released into the atmosphere. In essence, a solution indicated would be the establishment of a legal framework for the valorization of alternative energy sources from these deposits, by means of normative acts, issued at national level.

The treated and improved LFG represents a potential solution to the current energy crisis, felt both nationally and internationally, with the potential to reduce dependence on fossil fuel use. In the years ahead, these innovative technologies to produce purified LFG are expected to play a key role in shaping the future, because energy from renewable sources could contribute to alleviating current environmental and energy problems.

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