

INFLUENCE OF THE CHARACTERISTICS OF BIOGAS GENERATED IN THE LEATHER INDUSTRY ON COMBUSTION QUALITY

by

**Lucian MIHAESCU^a, Gheorghe LAZAROIU^b,
Gabriel P. NEGREANU^{a*}, and Ionel PISA^a**

^a Department of Technical Thermodynamics, Engines, Thermal and Refrigeration Equipments,
University "Politehnica" of Bucharest, Bucharest, Romania

^b Department of Energy Generation and Use, University "Politehnica" of Bucharest,
Bucharest, Romania

Original scientific paper
<https://doi.org/10.2298/TSC18S5349M>

The treatment of protein waste in the leather industry also includes transformation into biogas through anaerobic digestion. A pilot plant producing biogas with a 45-60% CH₄ concentration was designed for this purpose. Starting from primary experimental tests in a Bunsen burner, the paper presents the theoretical aspects of choosing the best combustion technique for this peculiar gaseous fuel in order to calculate the combustion velocity and checking the conditions for the flame stability. The applicability of the kinetic combustion (with pre-mix) will be attempted, because low combustion rates for this fuel also involve low air velocities. Diffusive combustion seems to not be affected by this impediment, as there is always an equality condition of flame velocity with burning speed, so a theoretical spot of a stable flame. After selecting the best combustion technique, a pilot burner will be designed and tested.

Key words: *biogass, combustion quality, flame stability, leather industry*

Introduction

The researches performed in the frame of a project [1] in order to neutralize the waste from tannery industry were focused on two main directions:

- combustion of animal fat in mixture with liquid fossil fuels, and
- anaerobic digestion of the protein with biogas generation.

The initial condition was the design and operation of a pilot digester with a discontinuous biogas generation rate. The biogas with a low calorific value (LCV) of 16300-21500 kJ/m_N³ was captured and stored, in order to be successfully burned in a Bunsen burner.

This paper purpose is to analyse the theoretical conditions and to present the results of biogas combustion. Due to the fact that the biogas resulted was in very small amount, without pressurized storage, the adopted solution was to use some similar gas compositions prepared by a specialised company. In papers [2-5] are presented biogas compositions with a CH₄ weight of about 55% and a CO₂ weight of 45%, similar to those obtained by anaerobic

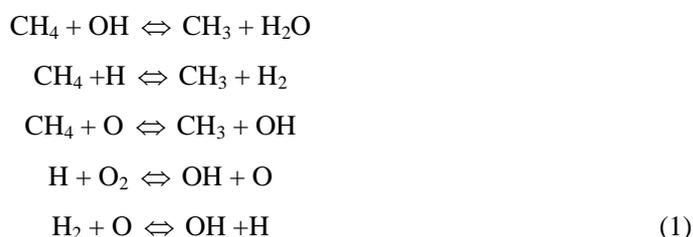
* Corresponding author, e-mail: gabriel.negreanu@upb.ro

digestion of the animal proteins from tannery industry. Even at vegetal biomass anaerobic digestion the CH₄ was in the range of 30-65%, the N₂ in the range of 2-17%, while the CO₂ was the rest.

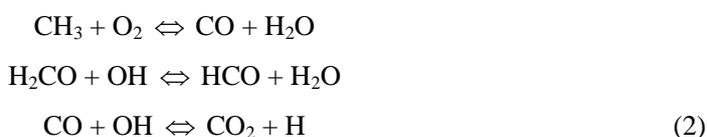
Characteristics of biogas combustion

The massive CO₂ fuel content leads to a decrease of the average temperature level of the flame. This fact contributes to the decrease of about 30% of the activation energy that will allow a more important role of the active reaction centres represented by the dissociation elements.

The first step of combustion is represented by following set of equations [6-8]:



Final step generates last combustion products [9]:



Before the first set of reaction, due to the high CO₂ concentration of the biogas, there is a presence of a conductivity heating zone, a chemically inactive area highlighted by the lack of specific luminosity of combustion. The size of this area depends directly on the concentration of the neutral CO₂ component. This chemically inactive area prefigures the initiation of ignition, and its excessive size disrupts both the ignition process and the flame propagation velocity.

The combustion dynamics changes spatially the fuel/air ratio so that the theoretical combustion rate in the stoichiometric concentration range remains only as a reference. For fuels rich in neutral gas (CO₂, N₂), where the flame initiation is disturbed, and the combustion rate will also suffer sensible decreases.

Biogas obtained through anaerobic digestions of animal proteins is fundamentally different from other low calorific gaseous fuels. The mentioned composition shows the absence of CO, which is dominant together to N₂ in the syngas obtained through solid fuels gasification [10-12].

The composition of the biogas was the following [13, 14]: CH₄ = 40-65%, CO₂ = 28-56%, N₂ = 3-6%, H₂ = 1-2%. This composition is defined by the ratio CH₄/CO₂, that, in this case, was in the range 0.7-2.32, while LCV was around 14400-21550 kJ/m_N³.

Before starting the theoretical and experimental analysis of biogas combustion, we compared the quality of this gas with a syngas obtained several years ago [13] by coal gasification. Its compositions is: CO₂ = 7.8-10.3%, C_mH_n = 0.1-0.3%, CH₄ = 2.5-3%, CO = 22-24%, H₂ = 13-18.3%, N₂ = 46.7-51.6%, while the LCV was in the range 5340-6038 kJ/m_N³. The purpose of this comparison is to emphasize the presence of H₂ and

CO previously mentioned, referring to its higher combustion rate. For this operation, the gasifier was of Lurgi type, and the burner was conceived for diffusive combustion, with axial slots for gas and swirled nozzles for air.

The real combustion rate, S_L^* [ms^{-1}], is corrected [14, 15] in relation to those resulted according to combustible elements S_L , because of diffusion of N_2 and CO_2 :

$$S_L^* = S_L (1 - 0.01\text{N}_2 - 0.012\text{CO}_2) \quad (3)$$

It is possible to admit that the massive presence of CO_2 in generated biogas represents a particular case. In fig. 1 is shown the links between the ratio CH_4/CO_2 and the relative combustion rate S_L^*/S and the LCV.

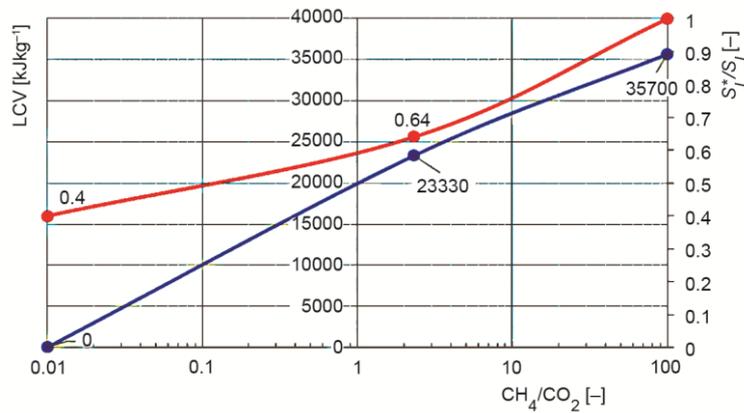


Figure 1. Biogas energy characteristics vs. CH_4/CO_2 ratio

In order to achieve the kinetic combustion, a stability condition is imposed by the low of combustion cone development [14]:

$$S_L^* = u \frac{R}{\sqrt{R^2 + h^2}} \quad (4)$$

where u [ms^{-1}] is the flowing velocity, R [m] – the radius of flow section, and h [m] – the flame height.

When the combustion rate S_L^* decreases, it should decrease also the flow velocity or the channel section. While the air-flow rate, V_a [m^3s^{-1}], is imposed by the combustion stoichiometry, the problem can be solved dividing the flow through many channels:

$$V_a = \sum \pi R^2 u \quad (5)$$

The parabolic profile of the flow velocity is given by:

$$\left(\frac{du}{dr} \right)_{r \rightarrow R} = \left(\frac{dS_L^*}{dr} \right)_{r \rightarrow R} \quad (6)$$

Non-swirled diffusive combustion imposes a stability condition much easier to achieve [14]:

$$u_m \leq S_T \quad (7)$$

where u_m [ms^{-1}] is the velocity in the middle of the jet, while S_T [ms^{-1}] represents the combustion rate in turbulent flows:

$$S_T = 0.18S_L^*(d_0 \cdot 10^{-2})^{0.26} \text{Re}^{0.24} \quad (8)$$

According to the distance x [m] from the flow section of diameter, d_0 , for a jet with initial exit velocity, u_0 , and the structure coefficient, a , the central velocity will be:

$$u_m = \frac{0.96u_0}{\frac{2ax}{d_0} + 0.29} \quad (9)$$

Combining eqs. (8) and (9), the maximal distance for the flame stability has been revealed:

$$x \leq 4.07 \frac{d_0^{0.75} \text{Re}^{-0.24}}{a} - 0.29d_0 \quad (10)$$

Experimental tests

Methodology

Due to the fact that the quantity of biogas generated by the mentioned digester was too small (an average value of $0.03 \text{ m}_N^3/\text{h}$ with large temporal fluctuations) it was impossible to use this gas for complex combustion experiments. In previous researches [15, 16] the collective who built the anaerobic digester has determined by mean of a chromatograph analyser the gas composition previously mentioned.

Taking into account that the average flow of the test burners used by us for gas combustion is about $0.7\text{-}2.0 \text{ m}_N^3/\text{h}$, we could not use the biogas generated by the digester and we ordered to Linde Gas, Romania to prepare three compositions of combustible gas, similar to the dispersion of biogas generated by the pilot anaerobic digester. The three qualities of gas were bottled in three cylindrical tanks, tab. 1.

Table 1. Composition of the three test gas

Quality/composition	CH ₄ [%]	CO ₂ [%]	H ₂ [%]	N ₂ [%]	LCV [kJm ⁻³ N ⁻¹]
Gas I *	52	45	1	2	18700
Gas II	60	32	1	7	21550
Gas III	40	56	1	3	14400

* Gas I represent the average quality

The tests were performed on an experimental stand for the determination of combustion characteristics of gaseous fuels, located in the Department of Technical Thermodynamics, Engines, Thermal and Refrigeration Equipments of University "Politehnica" of Bucharest.

The stand shown in fig. 2 comprises a non-cooled enclosure delimited by an optical viewing system. The tested burners are mounted at the bottom of the enclosure, the vertical

flame being ascending. Both gas-air pre-mixed through self-aspiration and diffusive burners with pressurized air supply can be used. In the left side of the picture it can be seen the air-flow control device.

In order to observe the combustion dynamics, an infrared video camera FLUKE 560 has been used, while the pollutant emissions were assessed by a gas analyser of type MULTILYZER.

The first set of tests was focused on *self-aspiration burner* equipped with a flame stabilizer (3 different types). In fig. 3 are presented the stabilizer with holes and stabilizer with slots:



Figure 2. Experimental stand equipped with a self-aspiration burner

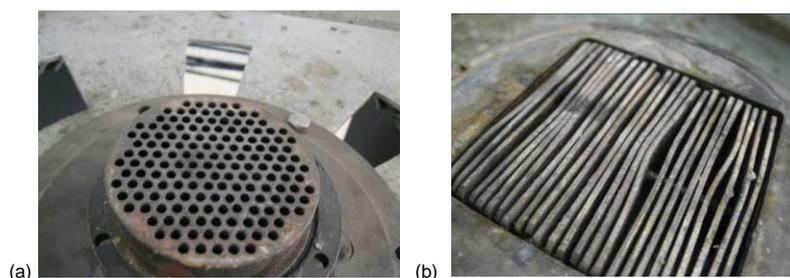


Figure 3. Flame stabilizers mounted over the self-aspiration burner; (a) stabilizer with holes (b) stabilizer with slots

The gas-flow rate was set to $1 \text{ m}^3_{\text{N}}/\text{h}$. The burner's thermal output for this gas-flow and quality is about $P_t = 5.4 \text{ kW}$. The gas-flow was imposed by the combustion rate because the increase of the flow over this value led to the flame extinguishment.

For the stabilizer with holes (having 160 holes with a hole diameter of 3.4 mm) the flow velocity for an air excess coefficient measured at the base of the flame $\lambda = 1.07$ was $u = 1.68 \text{ m/s}$. The flame stability requires that the condition $u \leq S_T^*$ must be assured.

The *diffusive flame burner* is shown in fig. 4. Fuel gas was admitted through a central hole of 5 mm in diameter and through four radial holes with a diameter of 0.5 mm. Air is blown through a peripheral crown that fits the fuel lance. The air had a velocity of 20-25 m/s. Tests were carried out with and without air swirling.



Figure 4. The diffusive combustion burner

For the gas-flow rate of $1 \text{ m}^3_{\text{N}}/\text{h}$, the outlet speed was 17 m/s, lower than that used for natural gas having a minimum value of 25 m/s.

Results of the experiments

For the self-aspiration burner operating with gas I, while combustion rate is 2.02 m/s the flame, shown in fig. 5, is stable, with a slight flame return trend. In fig. 6 is presented the flame of the burner equipped with a slots stabilizer.



Figure 5. Flame of the stabilizer with holes



Figure 6. Flame of the stabilizer with slots burner

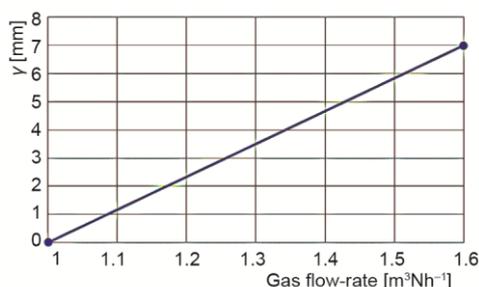


Figure 7. Dependence of the distance of flame formation vs. fuel-flow rate

An attempt to slowly increase the gas-flow rate (implicitly the thermal load) led to a flame detaching from the burner outlet section, with the occurrence of flame breakage. This tendency is shown in fig. 7, where is presented the dependence of the distance y from the burner outlet to the flame base vs. fuel-flow rate.

The temperature of the flame (measured with thermocouples) ranged between 1100-1200 °C depending on excess air coefficient. A qualitative image of the processes in the flame was obtained from the thermovision camera, as shown in fig. 8. The maximal temperature is recorded in the flame's core, based on dissociation processes revealed by eqs. (1) and (2).

Pollutant emissions recorded by the gas analyser were: $\text{NO}_x = 8\text{-}20$ ppm; CO varied between 3000 ppm in the stabilizer output section and 30 ppm at the end of the flame.

For the upper and lower limit fuels, namely gas II and gas III, see tab. 1, the burner's thermal power was maintained. Thus, the gas fuel flows were adjusted:

- for fuel gas II, $B_{\text{II}} = 0.87 \text{ m}_N^3/\text{h}$;
- for fuel gas III, $B_{\text{III}} = 1.3 \text{ m}_N^3/\text{h}$.

Experiments have shown a stable flame for both gas qualities. For fuel II (having a higher LCV) an increase in the base flame height to 50-60 mm (from about 30 mm for the fuel I) resulted. The increase in the temperature of the flame contour was about 30-50 °C.

For fuel III (with the lowest CH_4 content) there was a decrease in base flame height of about 7-14 mm, and a contour temperature of not more than 1065 °C. This fuel has led to an adequate combustion when the ceramic tunnel stabilizer was used, fig. 9.

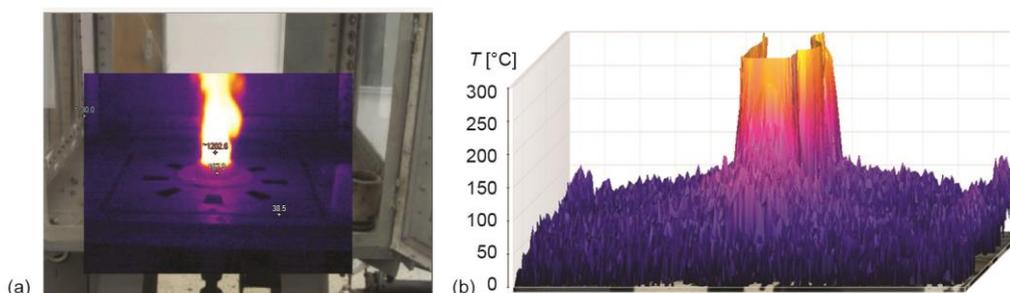


Figure 8 A qualitative image of the kinetic flame; (a) flame aspect, (b) temperatures in the flame core
 (for color image see journal web site)

The diffusive flame in three distinct cases (gas I, with non-swirled peripheral air and swirled peripheral air) are presented in figs. 10 and 11.

Thus, the flame has a considerable length and a high transparency rate, fig. 10. The use of swirled air contributes to a shorter and more stable flame, fig. 11.

The images revealed by the thermovision camera shows a peripheral cooling of the flame contour.

For atmospheric combustion, the excess air coefficient, λ [-], was in the range 1.2-1.23, while CO emission was registered at 590 ppm. The introduction of non-swirled peripheral air has led to an increase of the air excess coefficient λ from 1.2 to 1.32 at the base of the flame, with CO₂ emissions remaining at the same limit.



Figure 9. Flame aspect when using the ceramic stabilizer



Figure 10. Diffusive flame with non-swirled air



Figure 11. Diffusive flame with swirled air

The peripheral air swirling reduced the flame length to about 1/3 of the non-turbulent flame height and increased combustion intensity at the base of the flame so that the excess air coefficient, λ , could be lowered to 1.03-1.07, while the CO emission was in the range

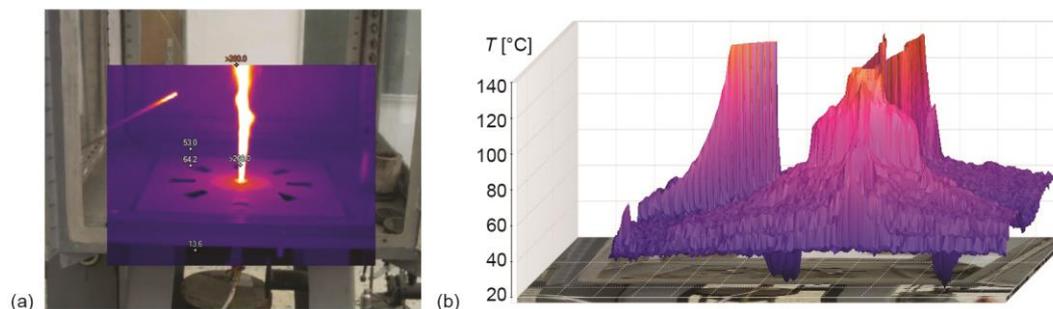


Figure 12. A qualitative image of the processes in the diffusive flame; (a) flame aspect, (b) temperatures in the flame (for color image see journal web site)

of 100-150 ppm. For all cases, there was a significant reduction in CO emission at the end of the flame, at average values of 40-45 ppm for combustion with atmospheric air, up to 5-15 ppm at swirled peripheral air combustion. The NO_x emission was below 20 ppm. The diffusive flames obtained were very transparent, with the average peripheral temperature below 450 °C. The results obtained with fuel III have shown that this gas quality burns properly, but the energy effect is far too low.

Conclusions

The use of prepared gaseous fuels in order to define the quality limits of biogas generated in the tannery industry allowed the following conclusions to be drawn.

- Both combustion technologies, kinetic and diffusive, can be used.
- Energetic gaseous fuel characteristics and performances are reduced when the CH_4 content drops below 50%.
- The low quality of the fuel gases with high CO_2 content and lack of CO imposed to quit the classic criterion (CH_4/CO) and adopting a new one, respectively (CH_4/CO_2), emphasizing the inhibitor role of carbon dioxide upon the flame development.
- The pollutant emissions (CO and NO_x) are below the limits required by the legislation.
- The fuel gas III with high CO_2 content has delayed the flame initialization and is characterized by the poorest energy effect.

The research concluded that is possible to use the similar gas like biogas generated by anaerobic digestion of waste-proteins from tanneries in the real range of composition and LCV, without modifications of industrial burners designed for natural gas combustion.

Acknowledgment

This work was supported by a grant of the Romanian National Authority For Scientific Research CNCS – UEFISCDI, project ID PN-II-PT-PCCA-2013-4-1017, *Green Tannery – Methods for Energetic Recovery of Biodegradable Wastes*.

Nomenclature

a – structure coefficient, [–]
 d_0 – hole diameter, [m]
 h – flame height, [m]
 P_t – thermal power, [kW]
 R – radius of flow section, [m]

S_L^* – real combustion rate, [ms^{-1}]
 S_L – theoretical combustion rate, [ms^{-1}]
 S_T – combustion rate in turbulent flows, [ms^{-1}]
 u – flowing velocity, [ms^{-1}]
 u_0 – initial exit velocity, [ms^{-1}]

u_m – velocity in the middle of the jet, [ms⁻¹]
 V_a – air-flow rate, [ms⁻¹]
 x – maximal distance for the flame stability, [m]
 y – distance from the burner outlet to the flame base [mm]

Greek symbol

λ – air excess coefficient [–]

Acronym

LCV – low calorific value, [kJkg⁻¹]

References

- [1] *** Green Tannery – Methods for Energetic Recovery of Biodegradable Wastes, project ID PN-II-PT-PCCA-2013-4-1017, UPB 2013-2017
- [2] Sutanto S., *et al.*, CO₂ Removal from Biogas with Supported Amine Sorbents: First Technical Evaluation Based on Experimental Data, *Separation and Purification Technology*, 184 (2017), Aug., pp. 12-25
- [3] Ward, A. J., *et al.*, Optimisation of the Anaerobic Digestion of Agricultural Resources, *Bioresource Technology*, 99 (2008), 17, pp. 7928-7940
- [4] Appels, L., *et al.*, Principles and Potential of Anaerobic Digestion of Waste-Activated Sludge, *Progress in Energy and Combustion Sciences*, 34 (2008), 6, pp. 755-781
- [5] Leclerc, C. A., Short Contact Time Catalytic Partial Oxidation of Biogas – A Comprehensive Study on CO₂ and N₂ Dilution, *Biomass and Bioenergy*, 63 (2014), Apr., pp. 58-63
- [6] Wang, W., Zhao, Y., Quantum Instanton Calculation of Rate Constant for CH₄ + OH → CH₃ + H₂O Reaction: Torsional Anharmonicity and Kinetic Isotope Effect, *The Journal of Chemical Physics*, 137 (2012), 21, ID 214306
- [7] Joseph, T., *et al.*, A New Potential Energy Surface for the CH₃+H₂↔CH₄+H Reaction: Calibration and Calculations of Rate Constants and Kinetic Isotope Effects by Variational Transition State Theory and Semiclassical Tunneling Calculations, *The Journal of Chemical Physics*, 87 (1998), ID 7036
- [8] Du, H., Hessler, J., Rate Coefficient for the Reaction H + O₂ → OH + O: Results at High Temperatures, 2000 to 5300 K, *The Journal of Chemical Physics*, 96 (1998), 2, 1077
- [9] Masunov, A. E., *et al.*, Chemical Reaction CO + OH → CO₂ + H Autocatalyzed by Carbon Dioxide: Quantum Chemical Study of the Potential Energy Surfaces, *J. Phys. Chem. A*, 120 (2016), 30, pp. 6023-6028
- [10] Angeladiki, I., Ahring, B. K., Anaerobic Thermophilic Digestion of Manure at Different Ammonia Loads – Effect of Temperature, *Water Research*, 28 (1994), 3, pp. 727-731
- [11] Monoli, B., *et al.*, Anaerobic Digestion of High-Nitrogen by Products in a Multiphase Process for Biogas Production, *Chemical Engineering Transactions*, 37 (2014), June, pp. 271-276
- [12] Yu, Liang, *et al.*, Two-Stage Anaerobic Digestion Systems where in One of the Stages Comprises a Two-Phase System, Patent Number: US 09567611, Patent Assignee: Washington State University, Official Gazette of the United States Patent and Trademark Office Patents, Pullman, Wash., USA, 2017
- [13] Panoiu, N., *et al.*, Modernization of Combustion Installations for Industrial Boilers (in Romanian), *Editura Tehnica*, Bucuresti, 1993
- [14] Stambuleanu, A., Industrial Flame (in Romanian), *Editura Tehnica*, Bucuresti, 1971
- [15] Mavrodin, M. E., Lazaroiu, Gh., Experimental Research on Combustion of Biogas Obtained through Anaerobic Fermentation of Tanneries Wastes^o, *U. P. B. Scientific Bulletin, Series B*, 80, (2018), 3, pp. 1454-2331
- [16] Craciun, M., *et al.*, Anaerobic Digestion of Tanneries Wastes, *Proceedings, 5th International Conference on Thermal Equipment, Renewable Energy and Rural Development TE-RE-RD 2016*, Golden Sands, Bulgaria, pp. 459-464