

Article

Substitution of Natural Gas by Biomethane: Operational Aspects in Industrial Equipment

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Abstract: Global gas markets are changing as natural gas (NG) is replaced by biomethane. Biomethane is produced by upgrading biogas, which can have a molar concentration of methane to over 98%. This renewable energy has been injected into the pipeline networks of NG, which offers the possibility to increase its usage in industrial and residential applications. However, the expectation of the increase in biomethane proportion on the NG grids could increase the fluctuations on the composition of the NG–biomethane mixture in amplitude and frequency. In this context, the injection of biomethane into the existing network of NG raises a discussion about the extent to which variations in gas quality will occur and what permissible limits should exist, as variations in combustion characteristics can affect the operation of the combustion processes, with consequences for consumers, distributors and gas producers. This study describes a gas quality analysis with regard to the use of biomethane in industrial equipment, mixed or not mixed with NG, taking into account the indicators for gas interchangeability and provides a discussion on the necessary gas quality level to be achieved or maintained for efficient combustion in equipment originally designed to operate with NG. NG and biomethane real data collected for 92 consecutive days in 2022 and provided by two different companies in Brazil were used for this study. It is shown that the maximum deviation of the Wobbe Index (WI) of 5%, which is allowed for industrial plants, does not work for the operation of furnaces at temperatures of 1200 °C or more. In addition, it is shown that the WI, as defined in relation to the calorific value of the fuel, may allow inappropriate substitution of fuel gases, which is likely to reduce the range of blending of biomethane in NG pipelines. The results can be assessed to analyze how the addition of biomethane to NG grids will impact the WI and the equipment operation parameters such as the air-to-gas ratio, products-to-gas ratio, adiabatic flame temperature and furnace temperature.

Keywords: biomethane; natural gas quality; industrial furnaces; industrial combustion



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1. Introduction

Biogas production has increased worldwide, stimulated by renewable energy policies as well as economic and environmental benefits. It is estimated that the global capacity of installed biogas increased from 65 GW in 2010 to 120 GW in 2019, 70% of which is in Europe [1]. This production is expected to grow until 2050, representing up to 27% of the global energy market [2].

Biogas comes from the digestion of organic residues such as agricultural, industrial and domestic waste, sewage sludge and others [3–8]; therefore, it can also be considered an environmentally friendly energy source. Biogas consists of three main components: methane (45~70%), carbon dioxide (24~40%) and nitrogen (1~17%) [9]. Secondary components of biogas include water vapor, oxygen, hydrogen sulfide, ammonia, carbon monoxide

and traces of halogenated hydrocarbons, siloxanes, toluene and others [2]. Hydrogen sulfide is the most corrosive element in biogas, which must be removed before utilizing it in industrial equipment [10]. The specific composition of biogas can vary during its production and is influenced by the type of feedstock used, the consumption of the feedstock in the reactor and the specific operating conditions of the bioreactor. Anaerobic digestion is the most widely used technology for biogas production [11] and has the potential to be an important process used by humankind for electricity and heat generation.

The energy density of biogas is relatively low compared to natural gas (NG). Since there are many processes that operate with NG, substituting NG for biogas in industrial equipment leads to a reduction in production capacity and stability. For this reason, intensive research has been conducted in recent years on technologies that improve the energy density of biogas. Biomethane or renewable natural gas (RNG) is the fuel obtained by upgrading and purifying biogas, and it can be used as an alternative for NG [3]. Since CO₂ is the most common non-combustible component of biogas, upgrading it to biomethane usually refers to the physicochemical removal of CO₂ or to the utilization of the CO₂ present on the biogas in other processes. During this conversion, most of the secondary components of biogas are also removed. Common methods of upgrading are classified as absorption, adsorption, membrane separation and cryogenic separation processes [1,2]. Another method of biomethane production is through the upgrading of syngas from biomass gasification [12].

As most of the biogas upgrading processes have reached maturity, the yield biomethane may not only be economically competitive but also can have over 95% of methane in its composition [9]. This means that biomethane can be distributed via the existing NG transmission pipeline networks on large scales without significant modifications on its infrastructure [9,13]. Europe is the leading producer of biomethane and it has been increasingly injecting it to the current NG network [11].

Recent developments on the use of biomethane as a substitute for NG have been studied extensively in recent years. Lee et al. [14] evaluated the emission characteristics of a stoichiometric natural gas engine fueled by compressed NG and biomethane and found that the thermal efficiency of engines operating with biomethane decreases by 2% when compared to NG. The authors also reported that the emissions remained below the requirements of the EURO-6 standards. Niamh [15] has made a techno-economic case study for biomethane injection and natural gas heavy goods vehicles and presented a methodology to identify the technical limitations of gas grid that imposes the amount of biomethane that can be injected into the grid, which the authors found to be between 34 and 40% of replacement. Adamo [16] has performed an economic analysis of biomethane production under different policy scenarios and provided policy implications that could enable further penetration of biomethane use as a substitute to natural gas in Italy. Mapelli et al. [17] has found that the use of biomethane to substitute NG in the steel industry as the most favorable when compared to green hydrogen in Italy. However, they mentioned that the substitution would be penalized by finding the amount of biomethane large enough to cover the demand of an ironmaking and steelmaking cycle.

Several studies have been conducted with the main objectives of analyzing fuel policies to promote biomethane development and economic studies to verify its financial feasibility [18–23]. There are three relevant issues that can be drawn from these studies: (a) there are still concerns about the quality and availability of biomethane in large scales; (b) most governments still need to improve policies that stimulate the use of biomethane to meet international agreements on greenhouse gas emissions; and (c) there is uncertainty related to the economic benefits of using biomethane due to possible impending cessation of government incentives.

Other studies have been made to provide an overview of the perspectives, potentials and challenges of biomethane in different countries. Examples include: description of biomass development and prospects in Europe [11], description of challenges in the use

of biomethane in the state of California, USA [7,8], overview of biogas application in Thailand [24] and description of the renewable energy potential in Brazil [25].

There are also some specific case studies, such as the estimation of land use efficiency for biomethane production in Sicily, Italy [6], development of a mathematical model for biomethane injection into the NG household grid in the UK [20] and evaluation of biogas use for electricity and heat production among small farmers in the city of Marechal Rondon, Brazil [26].

One application of biomethane in the industrial sector was the use of compressed biomethane as a substitute for liquefied petroleum gas (LPG) in the ceramics industry in Thailand [27]. Issues related to biogas production and biomethane upgrading have been discussed considering new technologies and the penetration of the renewable fuel in the country's energy balance [28,29].

Specific requirements for biomethane injection into NG pipeline networks, as well as standards for gas composition and combustion parameters, have been established by several countries, mainly in Europe [9,30]. The European regulations were divided into two parts, one for the automotive industry and the other for the gas grid injection: (a) the standard EN 16723—Part 1: specifications for biomethane for injection in the NG network; and (b) Part 2: automotive fuel specifications.

Most of the studies are focused on the economic aspects of the application of biomethane, and the environmental benefits and policies that would enable biomethane to be widely used. There are few studies that address the effects of operational quality due to fossil NG displacement by the introduction of biomethane in the industrial plants. Impacts on industrial process equipment and engines in the power sector have been assessed by Leicher et al. [31], who considered gas quality variations, mainly focusing on changes in gas markets due to the use of biomethane and hydrogen. Besides the impact in gas turbines and engines in the power plant sector, glass, ceramic and metals manufacturing are the most affected by changes in gas quality due to the requirement of high temperature processes. Another article of the same group of authors [32] described a rating of equipment for its sensitivity, based on the assessment of the impact of NG lower heating value (LHV) and Wobbe Index (WI) fluctuations on several industry processes. The article concluded that the consequences could be that these consumers need to invest in modern measurement and control systems to reduce the impact on the process. It is also worth mentioning the work on the simulation of the blending of fossil NG with hydrogen and biomethane in gas pipelines to evaluate the impact on gas combustion parameters, such as LHV and WI [33].

Fluctuations in gas quality can lead to poor equipment performance in industrial applications, such as inefficiencies, instabilities and unsafe combustion operation [34]. Advanced combustion systems have complex control technology to reduce emissions and improve efficiency. However, these systems could face significant operating issues with fluctuations in gas composition. Less complex designs may operate with large variations in gas quality, but are likely to have an impact on the emissions and process performance. Due to the expectation of the increase in biomethane proportion on the NG grids, variations on the composition of NG–biomethane mixtures in gas pipelines networks could increase in amplitude and frequency. Within this context, it becomes a relevant study that promotes an analysis about the extent to which variations in gas quality will occur and what the permissible limits are in terms of high temperature industrial equipment.

This paper presents a methodology to identify what should be the permissible limits of NG and biomethane blends for the case of furnaces with air-preheating. The analysis consists of identifying what is the mixture of NG and biomethane that complies with the WI requirement, which is usually used as the most important index to allow fuel interchangeability. The first part used NG and biomethane data collected during 92 consecutive days in 2022, provided by two different companies in Brazil. The data set is composed of gas compositions and heat contents. The impact of the WI (here defined by $WI = LHV/d^{0.5}$) on furnace operation parameters were analyzed. First, two types of high temperature furnaces and a low temperature furnace were analyzed. A ceramic tile furnace ($T = 1200\text{ }^{\circ}\text{C}$) with

air pre-heating of 220 °C and a glass furnace ($T = 1500$ °C) with air pre-heating of 1000 °C were taken as high temperature equipment. A fluidized bed sand dryer ($T = 600$ °C) was considered as a reference for a low temperature process. Usually, a variation on the WI range of 5% is permissible in industrial equipment [31,35,36]. However, the results of this paper show that this requirement is not sufficient for high temperature kilns. The second part uses average European data on the fuel gases and analyzes the emissivity of their mixtures and the requirements of biomethane injection parameters in the existing NG pipelines. The third part analyzes the permissible limits of blends between biomethane and NG for some European countries. This study provides a contribution to the discussion of NG quality variability and the procedures to determine combustion parameters, which were developed to analyze the consequences of using biomethane in high temperature industrial furnaces.

2. Materials and Methods

2.1. Lower Heating Value and Stoichiometric Air-to-Fuel Ratio

The dependence between the stoichiometric air-to-fuel ratio (RATIO) and the lower heating value (LHV) for a mixture of hydrocarbon fuels, such as natural gas and biomethane, is almost a straight line. This is the case for a data set provided by COMGÁS (Companhia de Gás de São Paulo, São Paulo, Brazil) for NG and by Gás Verde for biomethane. Gás Verde is a company based in Seropédica, in the state of Rio de Janeiro, which upgrades the biogas produced by the landfill that receives 10,000 tons/day of municipal waste from the city of Rio de Janeiro. In November 2022, the company's production capacity was 120,000 Nm³/day of biomethane and it can supply industries in the neighboring state of São Paulo. The data used was collected for 92 consecutive days, from 1 June to 31 August 2022.

In all calculations from this point on, the water vapor content of the air was assumed to be 1.5% (molar fraction). Figure 1 shows the variation of RATIO with the LHV for all available points. For such mixtures, the knowledge of the heating value is sufficient to determine the air-to-fuel ratio, which can be used to set the operating parameters of the furnace without knowing the exact chemical composition of the fuel being burned. The heating value of the gases was provided by COMGAS and Gás Verde.

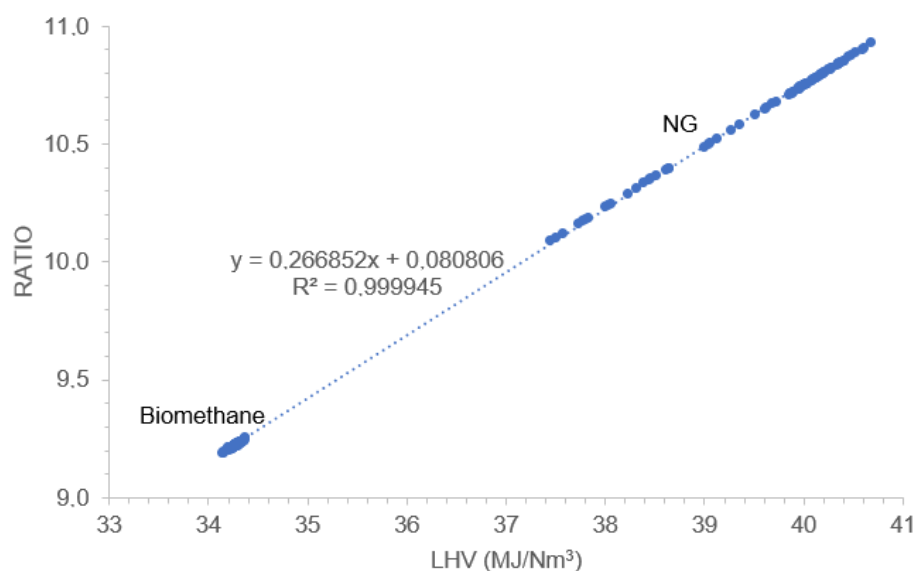


Figure 1. RATIO variation with LHV for NG and biomethane.

For the NG, the minimum LHV of the 92 data points was 37.45 MJ/Nm³ and the maximum value was 40.68 MJ/Nm³ (8.63% higher). The average was 39.63 MJ/Nm³. Also, for NG, the minimum RATIO was 10.09 and the maximum was 8.32% higher at 10.93. The average was 10.66.

Biomethane was more stable in terms of LHV and RATIO than NG, although it provides 86.5% of the heat input when both averaged. The LHV for biomethane was 34.14 MJ/Nm³ and the maximum was only 0.67% higher, 34.37 MJ/Nm³. The average LHV was 34.37 MJ/Nm³. In terms of RATIO, the minimum was 9.19 and the maximum was 9.25 (0.69% higher). The average was 9.23.

2.2. Wobbe Index

The most important parameter defining the interchangeability between two gases when using the same burner is the WI. The Index refers to the flow of thermal energy assuming that the conditions of the gas supply pressure and the diameter of the orifice through which the gas flows remain unchanged.

The WI is obtained from: Thermal energy flow rate (kJ/s) = Calorific value of the gas (kJ/Nm³) × Volume flow rate of the gas (Nm³/s), as follows:

$$Q = \Delta H_{gas} A k \sqrt{\frac{\Delta p}{\rho}} \quad (1)$$

where Q is the thermal energy rate, ΔH_{gas} is the calorific value of the gas, A is the cross-sectional area of the gas orifice in the burner, k is the orifice discharge coefficient, Δp is the gas supply gauge pressure, and ρ is the gas density. Rearranging, Equation (1) takes the following form:

$$Q = \Delta H_{gas} \frac{A k}{\sqrt{\rho_{air}}} \sqrt{\frac{\Delta p}{d}} \quad (2)$$

where ρ_{air} is the density of air and d is the density of the fuel gas in relation to air.

Wobbe defined his index as:

$$WI = \frac{\Delta H_{gas}}{\sqrt{d}} \quad (3)$$

such that:

$$Q = \frac{WI A k}{\sqrt{\rho_{air}}} \sqrt{\Delta p} \quad (4)$$

The calorific value of the fuel can be expressed either as higher heating value (HHV) or lower heating value (LHV). It has no influence on the choice, as the significance of the index lies in the percentage differences. In this article, the calculations are based on the LHV because the water contained in the combustion products remains as a gas on the flue gases.

Looking at the equation, it is evident that two different gases with the same WI will produce the same rate of thermal energy in a burner, as long as they have the same gauge pressure. One of the criteria adopted for two gases to be interchangeable is that their WI should be approximately the same, within a range of $\pm 5\%$ [31,35,36].

The same linear behavior as in Figure 1 does not occur for RATIO × WI because the denominator $d^{0.5}$ divides the LHV in the definition of WI. Figure 2 shows the dependence of the RATIO on the WI. The average CH₄ content of the 92 daily biomethane samples was 95.67%; the minimum was 94.84% and the maximum was 95.94%. Even with such a high CH₄ content, biomethane was not a suitable substitute for NG. The average WI was 7.25% lower for biomethane compared to NG; for the maximum and minimum WI's, the values were 7.88 and 6.82% lower, respectively. Therefore, biomethane is not interchangeable with NG under these conditions, as it is common industrial practice to allow a maximum deviation of $\pm 5\%$ WI.

2.3. Mixtures to Comply with WI Requirement

Mixtures were considered, defined by X biomethane + (1 − X) NG, where X is a mixture parameter. The discussion is based on the biomethane and NG given by their average concentrations for each of the 92 data points. Table 1 shows the volume compositions of NG, biomethane and the mixture corresponding to -5% of the original WI of NG (called

MIX). The value of X for the MIX was 0.698. This means that the energy input criterion of $WI_{MIX} > 0.95 WI_{NG}$ is not fulfilled for mixtures with a biomethane content of more than 69.8%. For more data on the average NG and biomethane and MIX used in the analysis, see Table 2.

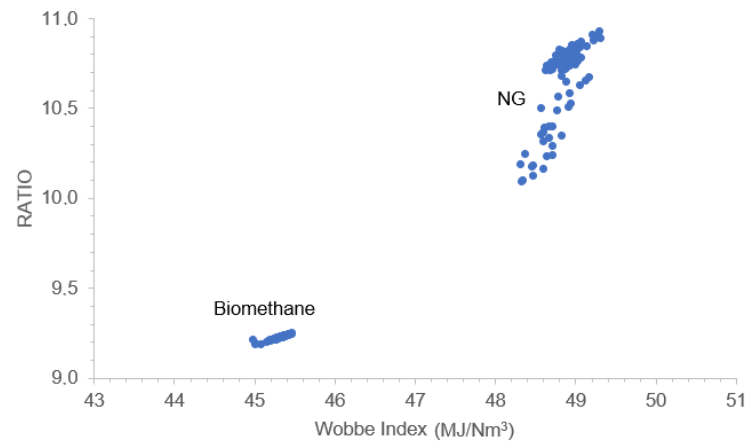


Figure 2. RATIO variation with WI for NG and biomethane.

Table 1. Volume compositions of average NG, average biomethane and MIX (X = 0.698).

	%N ₂	%CO ₂	%C1	%C2	%C3	%C4	%C5	%C6+	%H ₂ S	%O ₂
NG	0.649	1.959	85.14	8.231	3.222	0.698	0.070	0.025	-	-
Biomethane	3.83	-	95.67	-	-	-	-	-	0.027	0.47
MIX	2.868	0.592	92.488	2.487	0.974	0.211	0.021	0.008	0.019	0.331

Table 2. LHV, WI and RATIO of average NG, average biomethane and MIX (X = 0.698).

	LHV (MJ/Nm ³)	WI * (MJ/Nm ³)	RATIO **
NG	39.63	48.83	10.655
Biomethane	34.25	45.27	9.224
MIX	35.88	46.37	9.656

* Based on LHV; ** air with 1.5% moisture content.

The normalized excess of air or stoichiometric coefficient (SC) as a function of the oxygen concentration in dry basis in the combustion products is practically the same for NG, biomethane and MIX, as shown in Figure 3. The three curves are coincident. Therefore, the excess of air for any fuel mixture between NG and biomethane is given directly by the oxygen concentration, even though this concentration does not allow determination of which fuel is being burned. It should be mentioned that the graph in Figure 3 assumes a negligible CO concentration in the combustion products, which is not the case with a small excess of air. It should also be mentioned that industrial combustion processes are always set to operate with negligible CO emissions.

2.4. Economic Aspects of Substituting NG by Biomethane in Brazil

The price of NG depends on various unpredictable factors, such as political stability, trade agreements and the availability of NG. In Brazil, each state has specific policies and taxes related to the use and consumption of NG [37]. The NG cost from COMGÁS is related to the quantity consumed and it decreases from 1.04 USD/Nm³ for consumers that require less than 5 · 10⁴ m³/month to 0.76 USD/Nm³ for consumers using more than 2 × 10⁶ m³/month [38]. The average price of biomethane from Gas Verde is 0.8011 USD/Nm³ [39], however, it is still not injected into existing NG pipelines in Brazil and the final price increases due to the transportation cost.

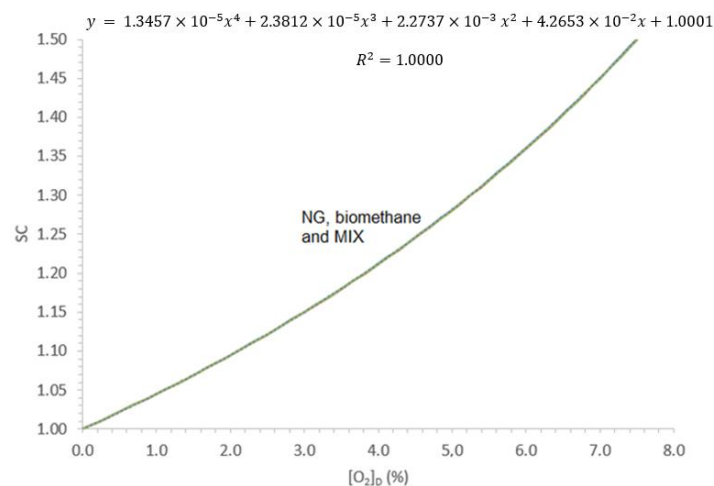


Figure 3. Normalized excess of air (SC) as function of the oxygen concentration in dry basis.

Figure 4 shows how these values relate to the heat required for a combustion process that runs for 730 h per month. The cost of the proposed mix is closer to biomethane than NG due to the higher proportion of biomethane in its composition. For consumers that require less than 5 MW and are located close to the producer, the cost of biomethane may be directly comparable to NG. However, for the majority of consumers, biomethane will be more expensive than NG. Forecasts indicate that the price of biomethane will decrease due to higher carbon allowance prices, while NG prices are expected to increase [40,41]. In the short term, a process using biomethane is not economically competitive compared to one that uses NG. In the medium and long term, industries will need to convert their plants to use pure biomethane to remain economically competitive.

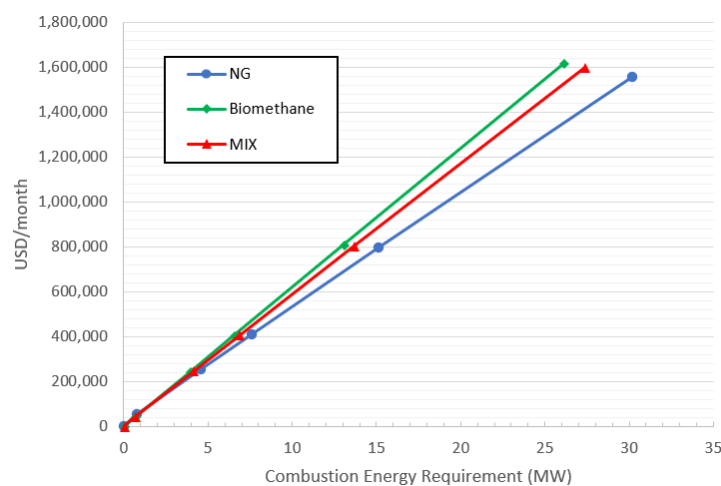


Figure 4. Costs of biomethane, mix and NG (Dec/2022).

2.5. Emissivity of the Combustion Products

Equation (5) was used to calculate the emissivity of CO₂ and H₂O. This equation was developed using regression analysis and agrees with the data of Hottel's emissivity charts. The emissivity of the gas mixture was calculated using Equation (6). The parameters a , b and d and the correction factors C_{CO_2} and C_{H_2O} are described by Mehrotra et al. [42]. Carbon monoxide and soot were not considered in the calculations.

$$\log \varepsilon_c = \frac{a + \sum_{i=1}^3 [b_i T^i + d_i (\log pL)^i]}{1 + \sum_{i=4}^6 [b_i T^{i-3} + d_i (\log pL)^{i-3}]} \quad (5)$$

$$\varepsilon_g = (\varepsilon_{CO_2} C_{CO_2} + \varepsilon_{H_2O} C_{H_2O}) \quad (6)$$

The temperature profile used to calculate the emissivity of the combustion gases was based on the results of a thermodynamic model applied to a billet reheating furnace in typical operation, as per the work of Mayr et al. [43,44]. The furnace has five sections: section one, soaking zone, sections two and three, heating zone, and section four and five, preheating zone.

3. Impacts on Equipment Operation

Now the effect on the furnace operation with the mixture 1 MIX = 0.698 biomethane + 0.302 NG (for which $W_{MIX} = 0.95 W_{NG}$) or with pure biomethane as a substitution for the corresponding pure NG is investigated.

Three cases are considered: (a) a high temperature furnace for the production of tiles (approximately 1200 °C), (b) a high temperature furnace for the production of glass (approximately 1500 °C) and (c) a low temperature furnace to provide hot gases for sand drying (approximately 500 °C). Standard heat balance methods [45] and a free chemical equilibrium program (GASEQ) [46] were used to calculate the combustion product enthalpies for different temperatures considering dissociation.

Adiabatic flame temperatures were calculated for both fuels. The differences between the temperatures for the same normalized excess of air are less than 6 °C for excesses of air above 1%. Therefore, both fuels can reach the same approximate adiabatic flame temperature and this is desired when substitution occurs.

3.1. Ceramic Tile Furnaces

An example of a kiln used for ceramic tile processing is a tunnel-like equipment composed of about 54 interconnected modules with a total length of 108 m. There are three main sections: (a) The ramp preheating section increases the temperature from 500 to 1200 °C and is responsible for the release of volatiles present in the ceramic tiles; this section represents between 20 and 25% of the furnace NG consumption; (b) the burning section, which keeps the temperature at 1200 ± 25 °C to harden the ceramic surface and it represents the consumption of 75 to 80% of the NG used in the furnace; and (c) the fast-cooling section, which cools the ceramic tiles.

The process heating is provided by a series of NG burners. The air flow rates are kept constant to keep the pressure, and the control parameter is the temperature. The temperature of each section is adjusted by varying the fuel flow rate according to the process temperature in each section. Air is used for the fast-cooling, which leaves the section at 220 °C and is used in the NG burners in the furnaces as pre-heated combustion.

Measurement of O₂ concentration is of great importance to ensure the overall performance of the heating process. This procedure allows a level of O₂ control sufficient to achieve complete oxidation of the organic matter present in the ceramic tiles and avoid defects in the final product. The furnace requires an O₂ concentration of more than 4% in dry basis in the entire extent of the furnace to prevent interference of the combustion gases with the product.

Figure 5 shows the O₂ concentration measurements obtained by two of the co-authors in a real ceramic furnace, on three different days: 15 January 2019 (initial condition), and 12 and 13 February 2019 (after adjusting the burner operation). The Figure also shows that, in green, the curve indicated for better operation of the mentioned equipment [47]. In fact, the comparison of production data for the two days of February showed a reduction in the specific consumption of NG of 12.1%.

Assuming a sudden, non-continuous change from NG by MIX, when the combustion control system is not designed to correct for the excess of air, the operator will be forced to change the excess of air manually to correct the heat input to achieve the operation required by the process. The latter is the most common condition, as advanced technology for this type of control is not available for most operating furnaces in Brazil.

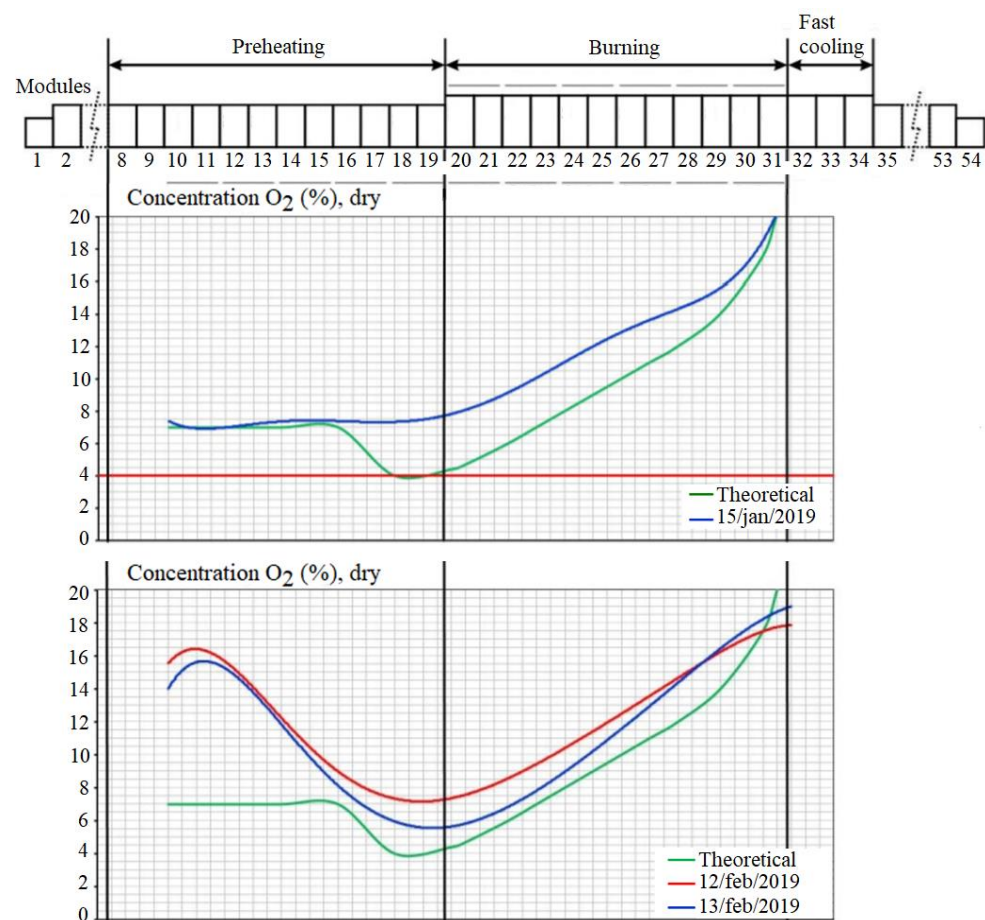


Figure 5. O₂ concentration along the length of a ceramic tile furnace.

In most cases, the furnace control parameter is temperature. While the air flow rate is kept constant, the control system increases the fuel flow rate until the desired temperature is reached.

This particular kiln operates with an average chamber temperature of 1200 °C and multiple burners, all configured to operate with NG. The preheated air enters the burners at a temperature of approximately 200 °C. The excess of air which corresponds to a minimum O₂ concentration of 4% is 21.2%.

If the two gases had the same WI, then $V_{NG}LHV_{NG}$ would be equal to $V_{MIX}LHV_{MIX}$ (recall the reasoning from Equations (1)–(4)), where V is the volumetric flow rate. In the case of $WI_{MIX} = 0.95 WI_{NG}$, $V_{MIX}LHV_{MIX}$ would be $0.95 V_{NG}LHV_{NG}$ in the case of a sudden change of fuels. Using the data from Table 2, $LHV_{NG}/LHV_{MIX} = 39.63/35.88 = 1.105 \sim 1.103 = 10.655/9.696 = \text{RATIO}_{NG}/\text{RATIO}_{MIX}$. Adding more fuel to meet the heat demand, the excess air is immediately reduced by 5%, i.e., at a constant air flow rate, the new SC is $0.95 \times 1.212 = 1.151$, which corresponds to an O₂ concentration of 3.0%, which is unsuitable for ceramic tile production.

An additional problem arises from the fact that the heat demand is not really proportional to the LHV. The variation of the combustion gas enthalpy with temperature for this situation is shown in Figure 6. Since the appropriate furnace atmosphere is 1200 °C, the amount of heat that needs to be supplied in the substitution is 378.4 kJ/mol, which is 10.64% higher than 342.0 kJ/mol. This reduces the normalized air-to-fuel ratio to 1.095, which corresponds to an O₂ concentration of 2%.

The difficulty of adjusting the heat input in a temperature-controlled constant air flow furnace for ceramic tiles becomes evident. Since the adjustment results in a lower excess of air when the gas is replaced by one with a lower WI, the oxygen profile reduces drastically.

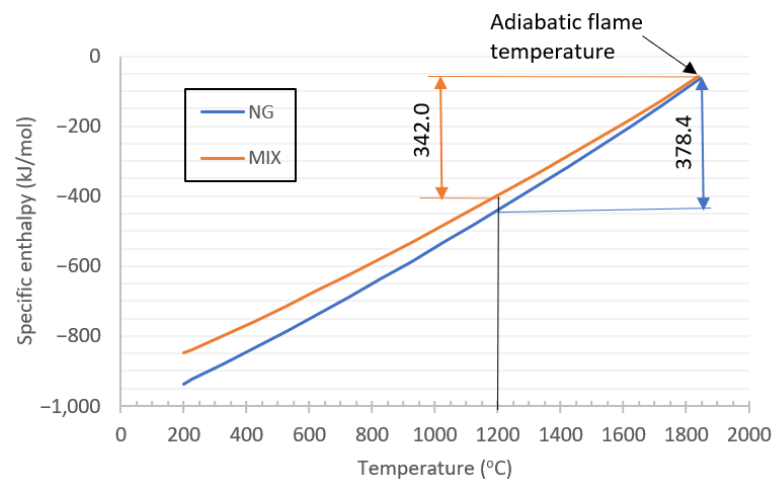


Figure 6. Enthalpy of combustion gases as function of temperature ($T_{\text{furnace}} = 1200\text{ }^{\circ}\text{C}$; $\text{SC} = 1.212$; $T_{\text{air}} = 200\text{ }^{\circ}\text{C}$).

3.2. Regenerative Glass Furnaces

Regenerative glass furnaces have two interconnected chambers. They contain refractory material and are called checkers. While the combustion gases flow through the checker in one chamber and enter the furnace, the checker in the other chamber is heated or regenerated with the hot exhaust gases that flow out. The reversal cycle takes about 20 min. Typical preheating temperatures of air are in the range of 900 to 1200 °C; temperatures inside the furnace are in the range of 1350 to 1500 °C [48]. The excess of air for combustion is usually in the range of 10 to 15% [49]. An illustrative figure of such equipment can be found in [50].

The case chosen for the investigation of the interchangeability of NG and MIX corresponds to an air preheating temperature of 1000 °C, an internal oven temperature of 1500 °C and an excess of air of 10%.

Figure 7 shows the dependence of the specific heat enthalpy of the combustion gases as function of temperature. In this case, the sudden change from NG to MIX requires an increase on heat input of 10.25% ($=560.3/508.2 - 1$), which takes the original normalized air-to-fuel ratio to under stoichiometric level. Therefore, substitution requires modern furnace control systems that are able to adjust operation to adequate levels of air-to-fuel ratios.

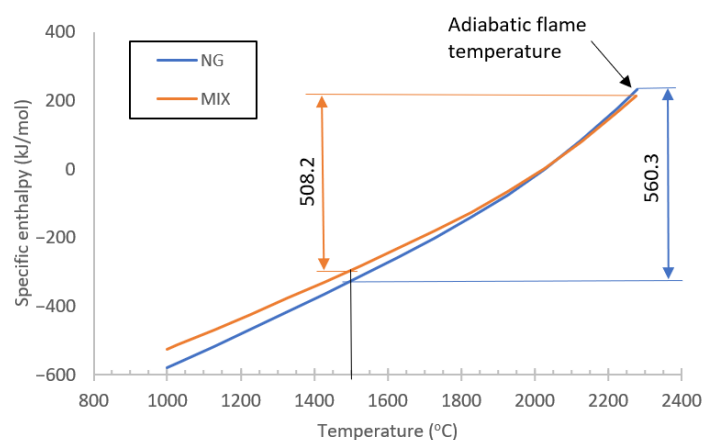


Figure 7. Enthalpy of combustion gases as function of temperature ($T_{\text{furnace}} = 1500\text{ }^{\circ}\text{C}$; $\text{SC} = 1.10$; $T_{\text{air}} = 1000\text{ }^{\circ}\text{C}$).

The content of this and the previous subsection are clear examples of how the range of the WI can be not narrow enough to achieve interchangeability. How narrow it needs to

be depends on a similar analysis for the specific process, at least in relation to the WI. The authors are aware that there are other indices and parameters that need to be taken into account to ensure full interchangeability.

3.3. Fluidized Bed Sand Dryers

The equipment considered here is an industrial sand dryer of the fluidized bed type (see Figure 8). Sand with a moisture content of generally 8 to 10% is the bubbling fluidized bed dryer where it is dried by a stream of hot combustion products in the range of 480 to 500 °C. To reach this temperature, dilution air is introduced into the combustion chamber. The operation of the equipment is controlled by the flow of sand and the flow of natural gas. Combustion air and dilution air flows remain practically constant. The total normalized air-to-fuel ratio and the absolute air-to-fuel ratio to reach 500 °C are listed in Table 3.

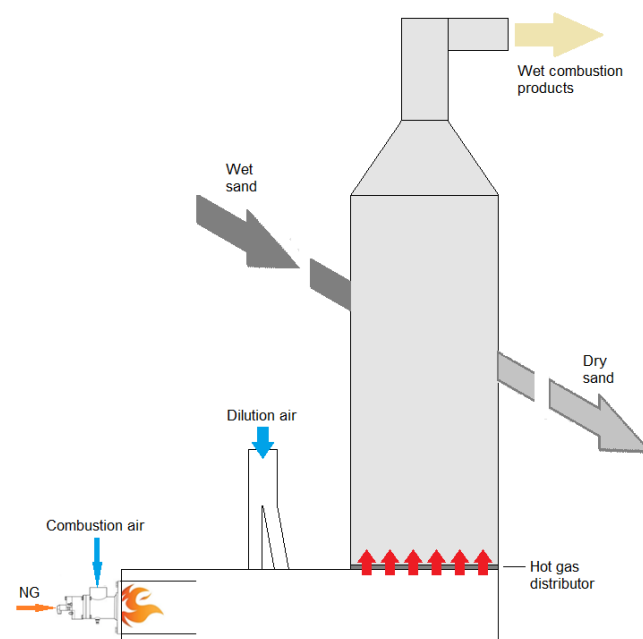


Figure 8. Fluidized bed type sand dryer (adapted from [39]).

Table 3. Air/fuel ratio, flue gas/fuel ratio for NG, biomethane and MIX ($X = 0.698$) to maintain 500 °C.

	Normalized Air/Fuel Ratio	Air/Fuel Ratio	Flue Gas/Fuel Ratio
NG	5.659	60.30	61.38
Biomethane	5.644	52.06	53.06
MIX	5.649	54.55	55.57

The data in Table 3 show that 60.30 Nm³ of air produces 61.38 Nm³ of flue gas at 500 °C when NG is burned. On the other hand, 60.30 Nm³ of air produces $60.30/52.06 \times 53.06$ Nm³ = 61.46 Nm³ of flue gas at the same temperature when burning biomethane. At a constant air flow rate, the flue gas flow rate therefore increases by only 0.13%. In this case, NG and pure biomethane are completely compatible given the low temperature required by the process.

4. Flue Gas Emissivity

Radiative heat transfer plays an important role in the operation of high temperature furnaces. In these furnaces, radiation heat transfer accounts for more than 90% of the total heat transfer, with the remainder being convective heat transfer [51]. Due to the fact that NG is mostly composed of hydrocarbons, the most significant contribution of radiative heat

transfer comes from carbon dioxide and water vapor [52]. Figure 9 shows the gas emissivity of two types of biomethane compared to Russian gas as a reference. The temperatures indicated at the top of the figure are averages for each section of the reheating furnace. There is no significant difference in the emissivity for NG and biomethane at the same gas temperature. The higher the temperature of the combustion products, the lower the emissivity of the gases.

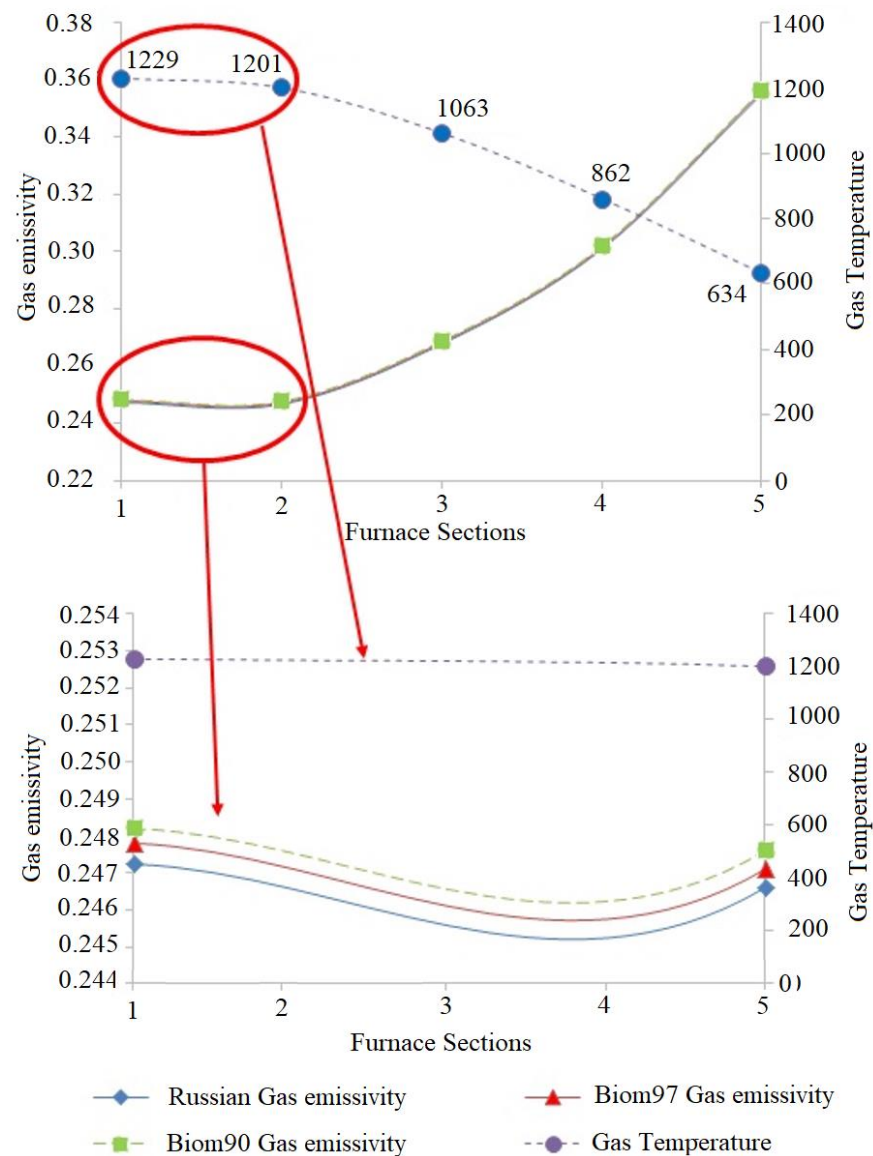


Figure 9. Gas emissivity per furnace section as a function of gas temperature.

The lower part of the Figure 9 shows an enlarged view of Sections 1 and 2. The emissivity of biomethane is slightly higher than that of NG. The emissivity of combustion products increases with the CO₂ content, as is the case with biomethane with 90% CH₄, as it has a higher CO₂ content. The data for Russian gas and biomethane are shown in Table 4. The table presents the chemical composition and combustion parameters for the different types of NG used. The data for fossil NG were taken from Altfeld et al. [53]. According to Khan et al. [9], biomethane chemical composition was considered from the requirements for injection into NG pipelines in Europe; the chemical composition of biomethane was considered based on the requirements for injection into NG pipelines in some European countries [54].

Table 4. Gas composition and combustion parameters of fossil NG and renewable NG.

	Russian Group H	G20 (Methane)	Biomethane 97%	Biomethane 96%	Biomethane 90%	Biomethane 85%
CH ₄ ^a	96.96	100.00	97.00	96.00	90.00	85.00
N ₂ ^a	0.86	0.00	0.58	0.78	3.00	8.00
CO ₂ ^a	0.18	0.00	2.26	3.01	6.00	6.00
C ₂ H ₆ ^a	1.37	0.00	0.00	0.00	0.00	0.00
C ₃ H ₈ ^a	0.45	0.00	0.00	0.00	0.00	0.00
n-C ₄ H ₁₀ ^a	0.15	0.00	0.00	0.00	0.00	0.00
n-C ₅ H ₁₂ ^a	0.02	0.00	0.00	0.00	0.00	0.00
n-C ₆ H ₁₄ ^a	0.01	0.00	0.00	0.00	0.00	0.00
O ₂ ^a	0.00	0.00	0.16	0.21	0.50	1.00
H ₂ ^a	0.00	0.00	0.00	0.00	0.50	0.00
Total ^a	100.00	100.00	100.00	100.00	100.00	100.00
HHV ^b	40.2	39.8	38.6	38.2	35.9	33.8
LHV ^b	36.3	35.8	34.8	34.4	32.3	33.8
ρ ^c	0.74	0.72	0.75	0.76	0.81	0.84
WI ^d	53.2	53.5	50.7	49.9	45.4	41.9

^a: mol %; ^b: MJ/Nm³; ^c: kg/Nm³ (density); ^d: MJ/Nm³ (WI based on HHV).

NG has a low carbon-to-hydrogen ratio and, therefore, tends to burn with a low-luminosity flame, because it produces few soot particles. However, there is another point of view regarding the CO₂ content in combustion products. Applications in high temperature furnaces, such as metals, minerals, ceramics and glass furnaces have NG burner designs to increase the flame luminosity through the production of soot. A luminous NG flame (yellow flame) is created by diffusion burners using techniques such as staged combustion. The first stage has a rich mixture close to the burner (fuel and primary air) to create soot particles based on the pyrolysis phenomenon and then, complete combustion takes place in a second stage through a secondary air injection. Emissions are kept below the standard values while luminous soot increases the flame emissivity and the heat transfer by radiation. Other techniques are injection of particles or oil into the flames, combustion with highly preheated air and/or combustion with highly preheated fuel [51,55–58]. Technologies to increase NG luminosity in high-temperature furnaces must be upgraded considering the advent of biomethane and hydrogen injection into NG pipeline networks, such as oxygen-enriched combustion. Several experimental studies have concluded that there is a notable increase in soot formation when the percentage of oxygen in the combustion air increases [54,59,60].

Biomethane fuels have the same carbon/hydrogen ratio as NG. Therefore, in principle, a flame produced by an industrial diffusion burner could have the same emissivity as the flame produced by NG. Nonetheless, if the CO₂ content in the biomethane is high, there will be a reduction in soot formation, decreasing the emissivity of the gas flame and flame luminosity. This phenomenon was studied, and it was concluded that soot formation is affected by dilution with CO₂ in methane diffusion flames. Other effects could be increased flame length and flame lift, depending on the amount of CO₂ content [61–64].

5. Biomethane Injection into Networks

The use of biomethane has several positive characteristics: reduction of the risk of exposure due to fossil natural gas regarding price and supply unpredictability; reduction in CO₂ gas emissions; decentralized energy production if biomethane small-scale units are available for injection into distribution systems; and biogas state of art upgrading technologies can produce biomethane that comply with the gas quality requirements for injection in pipelines and others.

In fact, biomethane should be a great opportunity to improve gas quality standards. This study investigated the blend of biomethane with fossil natural gas into pipeline networks with the purpose of reducing the WI variation range. The typical range of WI variation for some European countries is shown in Table 5. Those are measured values and the data was provided by the European network of transmission system operators for gas members [65] covering network points in several countries. The percentual change in

WI goes from 8% up to 15.8%, depending on the country. The level of disturbances that such variation can produce depend on the type of process, since some processes are more sensitive or not to WI variation, and of WI frequency distribution, which for most of the European countries is very broad, as raised by Aryal et al. [66]. However, as studied by Leichner et al. [31], certainly many processes are seriously affected.

Table 5. WI (based on HHV) measured ranges by country [54].

	NG Min WI	NG Max WI	WI Percent Range
	MJ/m ³ (25/0)	MJ/m ³ (25/0)	
Denmark	50.04	55.44	10.2%
Germany	49.32	54.00	9.1%
United Kingdom	49.86	54.00	8.0%
Italy	50.04	55.08	9.6%
Spain	50.40	56.52	11.4%
Netherlands	47.16	55.26	15.8%

As an example, if biomethane production is conducted in such a way that CH₄ content is between 97 and 98%, WI range will be from 50.7 to 51.6 MJ/m³, a variation of 1.8%. This gas injected into networks will reduce the existing WI range variation to an extent that will depend on the percentage of injection into the network. Two injection levels were simulated, the first considering the amount of biomethane required to achieve a gas mixed with 5% WI variation. The level of injection percentual is too high, for all countries, as indicated on Table 6. The second level of injection was defined at 20% for every country, and the gas mixed WI results are shown in Table 6. There was a significant reduction in WI variation for each country. The calculation was made considering the current flow of NG in the grids according to the energy balances of European Union Natural Gas Energy Balances [67] and does not account for fluid flow modeling in the NG pipeline or include multiple injection sources. Only an average calculation was made to demonstrate the real potential of reducing gas quality variation.

Table 6. Fossil natural gas mixed with biomethane—WI range.

	Biomethane Pipeline Injection		Current Average NG Pipeline Consumption	Blended NG + Biom. Min WI	Blended NG + Biom. Max WI	WI Current Percentage Variation Range	WI Blended NG + Biom. Percentage Variation Range
Units	%	TJ/Year	TJ/Year	MJ/m ³	MJ/m ³	%	%
Denmark	61%	61,000	100,000	50.44	53.10	10.2%	5.0%
	20%	20,000	100,000	50.17	54.67		8.2%
Germany	54%	1,258,200	2,330,000	50.07	52.70	9.1%	5.0%
	20%	466,000	2,330,000	49.60	53.52		7.3%
United Kingdom	47%	752,000	1,600,000	50.25	52.87	8.0%	5.0%
	20%	320,000	1,600,000	50.03	53.52		6.5%
Italy	58%	696,000	1,200,000	50.42	53.06	9.6%	5.0%
	20%	240,000	1,200,000	50.17	54.38		7.7%
Spain	66%	382,800	580,000	50.60	53.27	11.4%	5.0%
	20%	116,000	580,000	50.46	55.82		9.6%
Netherlands	76%	608,000	800,000	49.85	52.48	15.8%	5.0%
	20%	160,000	800,000	47.87	54.53		12.2%

One of the largest shares of biomethane in Europe is in Denmark, reaching over 10% of total gas consumption in 2019. It is estimated that 100% the gas consumption will be covered by biomethane in 2035 [68]. There are some barriers to connecting biogas plants to the gas grids to achieve this goal, such as high investments in the grids, costs for upgrading biogas to meet WI quality, gas compression costs and alternatives of using biomethane without the injection in the networks, thus avoiding the related high costs [66]. However, even with partial injection into the gas networks, if the biomethane has a small WI gap, there will be a great improvement to reduce variations in the quality of NG for end users. This is also in line with the NG quality requirements for internal combustion engine manufacturers, as they recommend limiting the WI range to 3 MJ/m³ and the maximum WI value to 53 MJ/m³ [69].

6. Conclusions

It has been commonly accepted that WI variations up to 5% are insensitive, and variations above that are sensitive to disturbances in operating factors such as efficiency, safety and product quality. However, this article has presented examples where this is far from adequate to ensure the proper operation of industrial equipment. In Section 2, a method was presented to determine the necessary mixtures to meet the requirements of WI. It was shown in Section 3 the impact on equipment operation and that the requirement of WI of up to 5% is insufficient for regenerative glass and ceramic tile kilns (high temperature processes), but sufficient for sand drying (a low temperature process). Section 4 has shown that there is no significant difference in the emissivity of NG and biomethane at the same gas temperature and Section 5 shows the proposed allowable blending of biomethane into the existing pipelines for some countries in Europe.

In the present article, three particular cases were considered (ceramic tile production, glass production, sand drying). The authors acknowledge that other major industries such as cement production, steel production and oil refining, which consume a large amount of NG and energy, could also be included. Specifically in steel production, reheating furnaces are combustion systems with three zones with different air-to-fuel ratios. One of the main problems of such furnaces is the formation of metal oxide deposits on the surface of the material, and when studying the change from NG to biomethane, this problem must also be taken into account. This is the subject of another study.

The issue of the quality of NG has also been considered. There are two main sides to it: consumers who have to deal with NG variations in quality and the resulting impact on their processes. They will have to invest in advanced automation and control technologies to increase efficiency and minimize productivity losses. To achieve this goal, it is important to immediately detect fluctuations in combustion parameters and make operational adjustments very quickly. It is therefore expected that a better understanding of the influence of combustion parameters and their interrelationships, as some presented in this article, will contribute to improving the logic of control systems. The other side is related to NG producers and distributors involved in the quality of the distributed gas. Biomethane is not only an opportunity to transition to a low-emission future as a complement to the use of existing gas infrastructure, but can also bring a change in gas quality. There are commercially developed and available biogas upgrading technologies that can produce biomethane with over 97% molar methane content. In addition, new technologies are being developed with emphasis on economic and environmental aspects. Biomethane can significantly contribute to stabilizing the energy supply and compensating for fluctuations in gas quality.

Regarding the specific conditions in Brazil, the biomethane considered in this study complies with the Brazilian regulation for the use of biomethane in the industry, which requires a minimum methane content of 95%. However, the results show that this value must be increased as more biomethane injection is added to NG grids to avoid operational consequences for consumers. European countries pioneering the use of biomethane require a minimum volumetric content of about 96–97%.

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Nomenclature

α	fuel molar fraction
C	a constant in Equation (4)
d	density in relation to air
ε_c	CO ₂ or H ₂ O emissivity
ε_g	flue gas emissivity
α_g	flue gas absorptivity
a	constant in Equation (5)
b	polynomial coefficient in Equation (5)
C_{CO_2}	pressure correction factor for CO ₂ emissivity
C_{H_2O}	pressure correction factor for H ₂ O emissivity
L	mean beam length
P	partial pressure
ρ	density
T	gas absolute temperature
X	mixing parameter between NG and biomethane

Subscripts

1	fuel number one
2	fuel number two
mix	fuel mixture

Abbreviations

G20	Pure methane
Biom85	Biomethane with 85% of CH ₄
Biom96	Biomethane with 96% of CH ₄
Biom97	Biomethane with 97% of CH ₄
Russian G	Natural gas from Russia Group H
NG	Fossil natural gas
RNG	Renewable natural gas or biomethane
MIX	Mixture of NG and biomethane
LHV	Lower heating value
RATIO	Stoichiometric air-to-fuel molar ratio
WI	Wobbe Index

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