Effects of inert gases on NOx formation in the conventional burner

I. Hudak*[,1], P. Belohradsky, P. Skryja

[1]Brno University of Technology, Faculty of Mechanical Engineering, Technicka 2, Brno 616 69, Czech Republic

Abstract

Low-calorific fuels have a unique composition and contain inert gases (such as N_2, CO_2) that are not involved in the combustion process directly, but on the other hand, they can reduce peak temperatures in the flame and therefore reduce the formation of NO_x emissions. The goal of this study is to prove whether the conventional burners, which were primarily designed for the combustion of natural gas, can be fired with alternative low-calorific fuels. Another objective is to check sustainability and economic balance.

Several types of low-calorific fuels were designed and calculated. The fuels will be then experimentally tested in the conventional burner. The selected low-calorific fuels are as similar as possible to the conventional low-calorific fuels. Lower heating values (LHV) of the designed fuels range between 6 – 20 MJ/m_N^3. As a reference fuel is used the transit natural gas with LHV around 36 MJ/m_N^3. The calculations were performed using CHEMCAD simulation software, with emphasis on lower and higher flammability limits and adiabatic flame temperature.

Introduction

In recent years, considerable emphasis has been placed on reducing hazardous emissions and decreasing fossil fuels consumption. The emissions reduction requirement can be achieved by the application of various strategies to the combustion processes, e.g. fuel or air staging, flameless combustion, etc. The requirement on improvement of the fuel conversion efficiency can be performed by e.g. recirculation of released heat back to incoming reactants using a recuperator or a regenerator. The improvement can also be achieved by the utilization of low-calorific fuels that are produced as by-products from e.g. distillation processes in petrochemical industry, landfills or pyrolysis processes.

The low-calorific fuels are characterized with fluctuating composition, calorific value and high inert content that may result in technical difficulties concerning their combustion, especially the burning stability. On the other hand, it is well known that lean combustion systems may suppress nitrogen oxides emissions (NO_x).

During last 20 years, many studies focused on the investigation of the composition of low-calorific gaseous fuels and the content of inert gases in noble gaseous fuels on combustion parameters were carried out. However, most of these studies were either purely numerical or the experiments were performed on laboratory-scale equipment focusing on the formation of NO_x and without the emphasis on thermal efficiency of the combustion process and stability of flame.

The effect of inerts on the combustion of methane has been investigated by several research groups. For instance, Li et al. [1] investigated the effect of nitrogen, carbon dioxide, argon and water vapor content on the formation of NO_x in methane flame. Zhao et al. [2] investigated the effect of steam addition in the methane flame. Several agents were chosen for the reduction of burning velocity of methane/air mixtures and the inhibition of flame by Fuss et al. [3]. The burning velocity of premixed flames was also studied by Liu et al. [4].

Methane was also used as the fuel in the experiments performed by Kobayashi et al. [5], [6]. In this study, premixed flames at high pressure and high temperature were diluted by superheated water vapour and CO_2. The effect of CO_2 dilution and pressure on the combustion of methane/air mixture was also analysed by Cohé et al. [7].

Another research group, Salvador et al. [8], performed experiments with natural gas burner and investigated the effect of addition of air and nitrogen as inert gas in the fuel.

Numerical investigation of the effects of syngas composition (H_2/CO/CH_4 mixture) and diluents N_2, H_2O and CO_2 on the structure and emission characteristics was reported by Giles et al. [9]. Other numerical studies of the effects of diluents on flame characteristics of syngas combustion were also performed by Park et al. [10], Glarborg et al. [11], and Chun et al. [12].

The $CH_4/O_2/NO_x$ system was used by Rasmussen et al. [13] to investigate sensitizing effects of NO_x on CH_4 oxidation.

The effects of N_2 addition to the combustion of Liquefied Petroleum Gas (LPG) was examined by Kumar et al. [14]. Research was focused on the soot formation, flame length and emission of NO_x, CO, CO_2.

The effect of water vapor injection was studied by Le Cong and Dagaut [15] and Mazas et al. [16]. Le Cong and Dagaut examined the effect of water vapor on the kinetics, especially on NO_x formation. Mazas et al. focused the research on combustion of lean, stoichiometric and rich fuels in terms of laminar burning velocity.

Most of numerical studies on laminar burning velocity uses GRI-MECH 3.0 mechanism [17]. For example, Boushaki et al. [18] studied laminar burning velocity of methane flame diluted by H_2 and steam.

* Corresponding author: hudak@upei.fme.vutbr.cz

Numerical results were compared with experimental data obtained on the slot burner.

The effect of steam dilution on combustion of natural gas and hydrogen was examined by Göke et al. [19]. The impact of steam on the burning velocity of ultra-wet methane/air/steam mixtures was examined by Albin et al. [20]. Other studies were focused on dilution effects of inert gases such as Ar, N_2, He and CO_2 [22], [23].

In laboratory scale were also conducted experiments on biogas. Zhen et al. [24], [25], [26] compared biogases with different compositions and also examined the effect of H_2 addition on the flame stability, flame temperature, transferred heat and emissions.

Tests on flameless combustion of biogas in lab-scale furnace were carried out by Hosseini et al. [27] with emphasis on flame temperature and species concentration through the flame. Same group investigated flameless combustion also numerically [28]. Tests on flameless combustion, dealing with temperature fields, species profiles and emissions were also performed by Colorado et al. [29].

Dai et al. [30] performed tests with six kinds of biogases on the reference test burner. Research was mainly focused on flame stability. Flame stability was also topic for Saediamiri et el. [31] who investigated effect of the swirl strength.

Laminar burning velocity methane and carbon dioxide mixtures were examined by Hinton et al. [32].

Summing up, the dilution of fuel has been investigated for a long time, however there is still lack of information about the experimental values of NO_x emissions, temperature profiles of diluted flame and heat flux distribution. Moreover, the majority of studies have been carried out under laboratory conditions. Therefore the results obtained by measurements at the large-scale testing facility might by markedly different.

ChemCad simulation

This work numerically investigate properties of selected low-calorific fuels. Results were obtained with CHEMCAD simulation software where all the fuels were designed and calculated. The flowsheet is shown in Fig. 1. Streams 1, 2, 3, 4, and 5 represents single component of the fuel used in the calculation. The components are mixed in mixers (Units 1 and 2) and together create the desired lean fuel (Stream 8). Afterwards the lean fuel is mixed in Unit 3 with the combustion air (Stream 6) and flows to the adiabatic combustion chamber (Unit 4). The chamber represents the equilibrium reactor that can simulate the combustion reactions. Flue gas (Stream 10) as a product is composed of water vapour, carbon dioxide and unreacted inert gases.. The Unit labelled with 5 is a back-feed controller that sets the amount of combustion air to keep the concentration of oxygen in the flue gas at 3 %vol., which corresponds to air surplus α=1,15.

Gases from three groups were selected for this study: biogas, producer gas and coke gas. Each gas is characterized by unique composition that results in different combustion characteristics such as flame stability, emissions or heat transfer rate. Gases are usually composed out of combustibles (such as CH_4, H_2 or CO) and inert gases (such as N_2, CO_2). The gases for the experiment will be supplied in cylinders, but due to the economic reasons it was decided that CH_4 will be substituted by the transit natural gas (NG) that is available at the facility. Higher Wobbe's number of CH_4 is ca. 50.7 MJ/m^3 while for NG it is ca. 50.5 MJ/m^3. Moreover, NG in the Czech Republic contains up to 97 %vol of CH_4 [33], therefore NG is an adequate substitute. Wobbe's number was calculated according to the formula given by Czech Technical Standard [34]:

$$W^0[t_1, V(t_2, p_2)] = \frac{\widetilde{H}_s^0[t_1, V(t_2, p_2)]}{\sqrt{d^0}}$$

where W is Wobbe's number [MJ/m^3], H is enthalpy based on volume basis [MJ/m^3], d is relative density[-].

Composition and parameters of selected gases are summarized in Table 1. As mentioned above, this study focuses on NG substitution in conventional burners by low-calorific fuels, therefore fuel No. (1) in the table is assigned to the NG, that is also considered as a reference gas.

Gases marked with labels (2) – (6) represent the group of biogases of different compositions. The difference among biogases (2) – (5) is in the amount of CO_2 in the fuel that ranges from 30 to 60 %vol. decreasing the concentration of flammable compound results in the decrease of Lower Heating Value (LHV) and Wobbe's number. It is known from the work [24] that mixtures with higher CO_2 are difficult to ignite. This could be caused by the change of the lower and higher flammability limits. They are shifted upwards, while the range remains the same and therefore it is difficult to stay within the limits during the ignition. Increased CO_2 concentration also lead to the change of the adiabatic flame temperature (AFT) that dropped almost by 200 °C, if the biogases with 30 and 60 %vol of CO_2 are compared. The biggest difference in AFT was obtained between NG and biogas (4) namely 273 °C.

The last biogas (6) contains approximately 50 %vol. of CH_4, 40 %vol of CO_2 and 10 %vol. of N_2. The amount of the inert gases in this fuel is same as in the biogas (4); LHV and flammability limits are also very similar, but Wobbe's number is slightly different. However, it is important to bear in mind that even though there is not a big difference between these fuels in terms of composition and LHV, flame characteristics, emissions and heat flux can be significantly different.

Producer gases differs from biogases especially by the amount and the composition of combustibles. Producer gas marked with (7) is characterized by high share of nitrogen (60 %vol), remaining 40 %vol is divided in between the combustible components (23 %vol. CO, 13 %vol. H_2 and 4 %vol. CH_4). Due to high ratio of inerts LHV is only 5.76 MJ/m$_N^3$, while Wobbe's number is 6.76 MJ/m$_N^3$. High ratio of inert nitrogen acting as the diluent causes that the adiabatic flame temperature is the lowest from all of simulated gases. On the other hand, this mixture is flammable in a wide range.

Total amount of inert gases for the second producer gas (8) is 24 %vol., therefore characteristics of this gas differs from the previous one. Even though there is 79 %vol. of combustibles, the LHV is only double, compared with (8) and so is the Wobbe's number. The main difference can be seen in the adiabatic flame temperature, where the increase by 250 °C can be observed.

The gas (9) has the highest concentration of hydrogen (60 %vol.) from all of the producer gases (22 %vol. of CH_4, 6 %vol. of CO and 14 %vol. CO_2). Compared with NG the LHV is about three times lower, lower flammable limit is very similar, but higher flammable limit is higher (40.51 %vol.). On the other hand, the adiabatic temperature is slightly higher than the adiabatic temperature of NG.

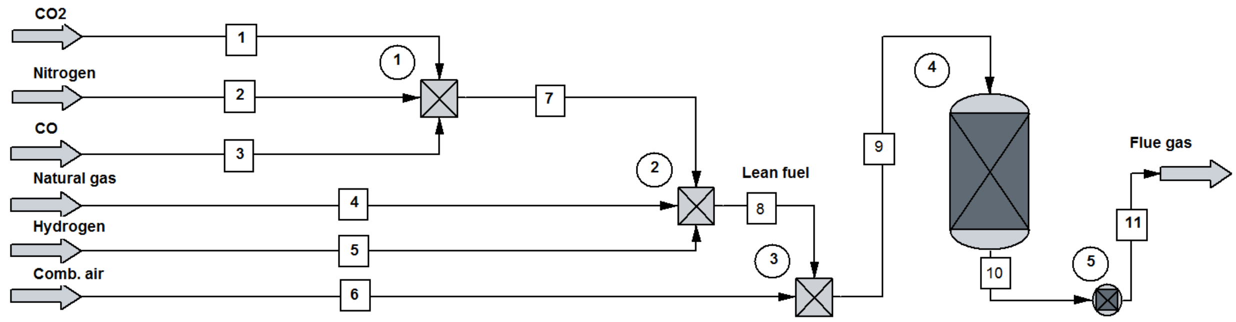

Fig. 1: Scheme of process in CHEMCAD simulation software.

The last category of low-calorific fuels included two gases obtained from coal. In the first fuel (10), almost no inert gases and a trace amount of nitrogen coming from NG are contained. On one hand, due to high hydrogen concentration Wobbe's number is relatively high (41.56 MJ/m_N^3) compared with other gases, but on the other hand LHV is lower (19.8 MJ/m_N^3) than LHV of NG. The adiabatic flame temperature for this fuel is 1847 °C which is the highest value from all gases.

The second coal gas (11) contains 24 %vol. of NG and 16 %vol. of inert gases. Thus the adiabatic flame temperature, LHV and Wobbe's number are lower than for the fuel (10).

Future work – experimental study

Combustion tests will be carried out at the burners testing facility, shown in Fig. 2. Facility enables to carry out the combustion tests and to collect experimental data for further assessment and verification of numerical simulation results. Burner capacity will be set to 500 kW and the investigated parameters will include NO_x and CO emissions, flue gas temperature, heat flux to the wall of the combustion chamber as a measure of thermal efficiency, distribution of in-flame temperatures in the horizontal symmetry plane of the combustion chamber, and the stability, shape and dimensions of the flame. Moreover, the noble fuel will be also diluted by admixing inert gases like nitrogen, carbon dioxide, and saturated steam. The effect of these inerts on flame properties will be investigated, too.

The key apparatus of the facility is a two-shell horizontal water-cooled combustion chamber with the inner diameter of 1 m and the length of 4 m. The front side and the rear side of the chamber are insulated with the high temperature fibrous lining with the thickness of 100 mm. The cooling shell of the combustion chamber is divided into seven individual sections with independent supply of cooling water. Each section is equipped with sensors for the measurement of flow rate, inlet and outlet temperature of cooling water. Water flow rate is measured by turbine flow meters, and inlet and outlet temperatures are measured by resistance thermometers placed in the steel sheath.

The combustion chamber is equipped with eight inspection windows along the cylindrical part and two more inspection windows on the rear side opposite the burner which allow observing of the flame on the burner. The windows can be also used for the installation of additional measurement instrumentation, e.g. thermocouples, heat flux probe, etc.

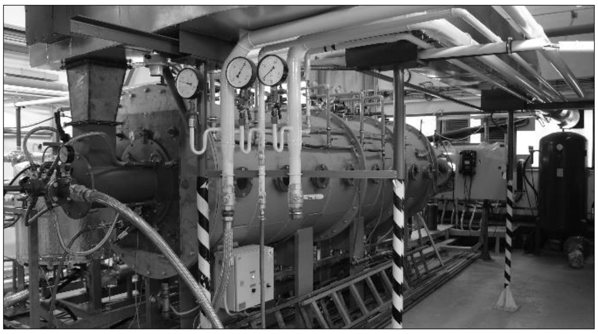

Fig.2. The burners testing facility.

Flue gas is exhausted from the combustion chamber through the flue gas stack. There are three measurement and sampling spots for measuring pressure in the combustion chamber, flue gas temperature and flue gas composition. The flue gas analysis and flue gas temperature measurement are provided by the flue gas analyser TESTO 350-XL analyser. The analysis box is equipped with electrochemical sensors for real time measurement of O_2, CO, CO_2, NO and NO_2

concentrations in dry flue gas. The flue gas temperature is measured using the K-type thermocouple. The negative pressure in the combustion chamber, which can be as low as -400 Pa, is created using an ejector located in the lower part of the stack.

Combustion air is supplied to the front of combustion chamber using high-pressure fan that is equipped with the frequency converter. The maximum fan output is approximately 4500 m_N^3/h with maximum overpressure of 4 kPa.

The testing facility is equipped with sophisticated data collection and safety system. The system for data collection enables automatic data collection every second or two minutes as well as data collection upon operator's request. Collected data include flow rates of combustion air, fuel and cooling water, temperatures of combustion air, fuel and cooling water, pressures in intake pipes and composition and temperature of flue gas. The safety system ensures safe and reliable operation of testing facility using information from sensors, e.g. it prevents preheating of cooling water and flame blow-off.

Table 1
Composition and parameters of selected fuels and flue gas.

Fuel component	**Component concentration [%vol]**										
	1	2	3	4	5	6	7	8	9	10	11
Hydrogen	0	0	0	0	0	0	13.042	37.999	58.001	60.040	60.013
Methane	97.101	68.04	57.992	48.551	38.840	48.592	3.891	9.710	21.517	33.95	23.304
Ethane	1.590	1.113	0.954	0.795	0.646	0.797	0.064	0.159	0.350	0.556	0.381
Propane	0.362	0.254	0.21	0.181	0.142	0.184	0.015	0.036	0.081	0.126	0.087
N-Butane	0.057	0.040	0.034	0.028	0.0220	0.029	0.003	0.006	0.013	0.02	0.014
Isobutane	0.061	0.043	0.367	0.031	0.024	0.031	0.003	0.006	0.013	0.021	0.015
Pentane	0.010	0.007	0.006	0.005	0.004	0.005	0.001	0.001	0.002	0.004	0.002
Hexane	0.010	0.007	0.006	0.005	0.004	0.005	0.001	0.001	0.002	0.004	0.002
Carbon Monoxide	0	0	0	0	0	0	23.044	28.107	5.999	5.001	0.024
Carbon Dioxide	0.100	30.001	40.005	50.051	60.03	40.052	0.004	23.911	14.022	0.032	8.002
Nitrogen	0.709	0.496	0.425	0.355	0.280	10.355	59.943	0.071	0.156	0.252	8.175
Fuel Parameters											
LHV [MJ/m_N^3]	36.3	28.2	21.78	18.15	14.52	18.15	5.76	11.27	15	19.79	16.19
HHV [MJ/m_N^3]	40.3	25.41	24.17	20.14	16.12	20.15	6.17	12.42	17.02	22.37	18.34
LFL [%]	4.95	6.92	7.98	9.43	11.51	9.43	15.39	7.14	5.11	4.45	4.86
HFL [%]	15.01	20.14	22.73	26.1	30.62	26.09	73.94	55.64	40.51	31.2	38.52
Wobbe [MJ/m_N^3]	53.3	30.49	24.79	19.7	15.09	20.25	6.76	14.66	25.76	41.56	31.74
Density [kg/m^3]	0.738	1.106	1.164	1.351	1.473	1.279	1.079	0.928	0.564	0.374	0.431
AFT [°C]	1787	1703	1660	1604	1514	1615	1503	1765	1794	1847	1841
Flue gas component											
Carbon Dioxide	8.138	12.652	14.718	18.049	13.264	11.180	16.466	8.338	6.181	6.017	12.652
Oxygen	2.999	3	3	2.999	3	3	2.997	3	3	3	3
Water	16.957	15.978	15.529	14.806	15.53	9.382	16.232	20.938	20.672	20.887	15.978
Nitrogen	71.051	67.555	65.959	63.383	67.412	75.822	63.539	66.918	69.312	69.281	67.555
Argon	0.855	0.815	0.794	0.763	0.794	0.616	0.766	0.806	0.835	0.816	0.815

Conclusions

A numerical study of low-calorific fuels is presented in this work. Several fuels have been selected and simulated in CHEMCAD simulation software.

The goal of this study was to understand the behaviour of low-calorific fuels and find out the dependence in concentrations of flammable compounds and inert gases. Increasing ratio of inert gases in the fuel results in decrease of lower heating value and Wobbe's number. Higher amount of inert gases also change the flammable limits, i.e. the range where a fuel can be ignited is much wider than the range for natural gas. The difference between the adiabatic flame temperatures was also observed. Next the higher amount of incombustible substances in the fuel acts as a diluent which absorbs significant amount of produced heat and is wasted by the flue gas.

In the experimental part, which will follow this study, the effect of low-calorific fuels on the pollutant emissions, flue gas temperature, heat flux to the wall of the combustion chamber as a measure of thermal efficiency, distribution of in-flame temperatures in the horizontal symmetry plane of the combustion chamber, and the stability, shape and dimensions of the flame will be investigated.

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