

## EXPERIMENTAL STUDY ON COMBUSTION OF CH<sub>4</sub>/NH<sub>3</sub> FUEL BLENDS IN AN INDUSTRIAL FURNACE OPERATED IN FLAMELESS CONDITIONS

by

**Rafal SLEFARSKI\*, Pawel CZYZEWSKI, and Michal GOLEBIEWSKI**

Faculty of Environmental Engineering and Energy,  
Poznan University of Technology, Poznan, Poland

Original scientific paper  
<https://doi.org/10.2298/TSCI200401282S>

*This paper presents the results of an experimental study on the combustion process of methane mixed with NH<sub>3</sub> in flameless mode. At a time of striving for CO<sub>2</sub>-free power, NH<sub>3</sub> became a potential energy storage carrier fuel from renewable sources. Flameless combustion features low emissions and is a very efficient technology used in the power sector, as well as steel production, ceramics, etc. Industrial furnaces were tested in the context of pure methane combustion with an addition of NH<sub>3</sub>, up to 5%. Flameless combustion conditions were achieved with a regenerative gas burner system (high regenerative system). The burner consists of four ceramic regenerators allowing for continuous preheating of air, even up to 50 K lower than the temperature of the combustion chamber wall. Constant power of the introduced fuel was kept at 150 kW and the fuel-air equivalence ratio ranged from 0.75 to 0.95. The results have shown a growth of molar fraction of NO in flue gases when NH<sub>3</sub> content in the fuel rose. The increase is more significant for the tests with a higher amount of oxygen in the combustion chamber (a lower fuel-air equivalence ratio). An addition of 5% of NH<sub>3</sub> into the fuel caused an emission of NO at the levels of 113 ppmv and 462 ppmv (calculated to O<sub>2</sub> = 0%), respectively for low and high fuel-air equivalence ratios.*

*The calculated conversion factor of NH<sub>3</sub> to fuel NO has shown extremely low values, equal to 2% ( $\phi = 0.95$ ) and 8.4% ( $\phi = 0.75$ ), which indeed confirmed that NH<sub>3</sub> can be burned with low emissions in flameless combustion technology.*

Keywords: NH<sub>3</sub>, flameless combustion, NH<sub>3</sub> energy storage, fuel NO, CH<sub>4</sub>/NH<sub>3</sub> combustion process.

### Introduction

The current climate challenges [1] trigger major transformations to low-carbon energy generation in the industrial and the energy sectors. According to the current state of the art, these challenges have to be achieved through the extensive use of RES and the use of modern energy storage methods. Currently, the most common energy storage method is pumped-storage hydroelectricity [2]. However, it requires the exclusion of very large areas of land and therefore has a negative impact on the environment. Consequently, a wider use of this method is limited and unable to meet the growing demand for energy storage volume of

\*Corresponding author, e-mail: rafal.slefarski@put.poznan.pl

energy sector, induced by the increasing share of unstable energy surplus from the sun and wind power plants [3]. Many technologies for electricity surplus storage in the form of energy carriers, such as hydrogen, synthetic methane, heat or direct accumulation in batteries, are proposed. Not only do they differ in efficiency, but also in the density of the stored energy, which significantly limits their potential use [4, 5]. One of the most widely studied storage methods is the production of renewable hydrogen from green energy sources via electrolysis of water. It can be used in industrial combustion systems, energetic engines or fuel cells [6]. However, hydrogen is a low energy density fuel, problematic in storage and transportation. Therefore, special emphasis is placed on its derived components such as synthesis gases [7] or NH<sub>3</sub> [6] as energy storage carriers. The NH<sub>3</sub> is used as a component for many industrial products: as a refrigerant in industrial coolers, as well as fertilizers in agriculture. In the energy sector it is known as NO<sub>x</sub> – a reduction agent used in selective non-catalytic reduction technologies. Such widespread use means that the processes of NH<sub>3</sub> production, transportation and storage are well known and do facilitate its use as a carbon-free fuel. However, the use of NH<sub>3</sub> as fuel involves several difficulties resulting from its physicochemical properties. Compared to hydrocarbon fuels, NH<sub>3</sub> has narrower flammability limit, higher ignition temperature, lower flame temperature and lower burning velocity. Combustion of NH<sub>3</sub>/air mixtures also delivers smaller amounts of energy released from oxidation reactions and causes a reduction of radiation heat transfer from the flame (lack of CO<sub>2</sub> in combustion products) [8]. Another negative aspect of NH<sub>3</sub> use as fuel is NO emissions. Due to the nitrogen bound in the NH<sub>3</sub> structure its oxidation involves high risk of NO<sub>x</sub> emissions (fuel mechanism NO<sub>x</sub>), significantly exceeding the current legal limits [9, 10]. However, the necessity of electricity and heat production decarbonisation, resulted in the development of technologies for the utilization of NH<sub>3</sub> and its mixtures with other fuels in reciprocating gas engines or gas turbines [11-13]. Flameless combustion, which is based on exhaust gas recirculation in which NO<sub>x</sub> formation is suppressed without compromising the thermal efficiency, is the potential technology tackling the NO<sub>x</sub> emission problem. The exhaust gas is recirculated and mixed with fresh air before the reaction. As a result, the peak combustion temperature is reduced. However, if exceeding certain limits, it can also lead to unstable combustion and blow off [14].

The potential use of NH<sub>3</sub> as a fuel has also been tested for energy generation in industrial furnaces and boilers. Yamamoto *et al.* [15] presented the possibility of co-combustion of pulverized coal with a percentage of NH<sub>3</sub> blending (max. 20% LHV basis), using a horizontal test furnace with a single burner. They observed that the maximum NO<sub>x</sub> emission did not increase with the co-injection of NH<sub>3</sub> into the furnace at ratios of less than 10% of the total LHV of the fuel. Szewczyk *et al.* [16] studied an experimentally modified flameless combustion technology (ZVC technology) for low, medium and high calorific fuels doped with NH<sub>3</sub>. They showed that the conversion of NH<sub>3</sub> to NOs strongly depends on the amount of oxygen in the zone where the fuel is injected. Decreasing the oxygen content caused a decrease of NO formation for both syngas and hydrocarbon fuels. Finally, Sorrentino *et al.* [17] presented the results of a study of NH<sub>3</sub> combustion under highly preheated and diluted conditions in a cyclonic burner. They showed that internal recirculation induced by the cyclonic burner ensured the stabilization of the flameless combustion process, even for pure NH<sub>3</sub> with extremely low NO<sub>x</sub> levels (below 100 ppmv). Sullivan *et al.* [18] have observed a very important effect on NH<sub>3</sub> decomposition in non-premixed methane-air flames: they noticed that NH<sub>3</sub> is decomposed and NO is formed mainly on the border between the fuel rich and the fuel lean mixture. In case of flameless conditions, where high injection velocity and strong internal recirculation is observed, the combustible mixture is lean. The rise of air con-

tent converts the combustion mixture into leaner fuel and the abundant oxygen intensifies the conversion of NH<sub>3</sub> to NO. Zieba *et al.* [19] shows that NH<sub>3</sub> decomposition in free jet fuels is divided into two stages: before and after the ignition. In the pre-ignition zone, methane slowly converts to methyl radicals and consumes most of the H, O, and OH radicals found in the jet. At the same time, NH<sub>3</sub> conversion does not proceed because of the lack of radicals able to react with NH<sub>3</sub> in the low temperature range. When the fuel ignites, NH<sub>3</sub> decomposition starts very rapidly. Because fuel is mixed with a combustion air jet and the conditions are over stoichiometric almost all of the NH<sub>3</sub> is converted to NO.

This paper concerns itself with the research on the co-firing of methane with an NH<sub>3</sub> in flameless combustion technology in an industrial combustion chamber equipped with an industrial high regenerative system (HRS) burner. Flameless combustion technology has shown great potential in the context of increasing the efficiency of energy systems, while significantly reducing the emissions of NO<sub>x</sub>. It is also a potential technology for high efficiency utilization of non-standard low calorific gaseous fuels. The goal of this paper was to study the influence of physicochemical properties of fuels mixtures and the exploitation parameters on the operating features of industrial furnaces operating in flameless technology. Pure methane and three mixed levels of NH<sub>3</sub>, up to 5% in volume, have been experimentally tested and compared.

### Test rig

Research of methane-NH<sub>3</sub> blends' combustion process in an oxidizer at a temperature higher than the self-ignition temperature of the combustible mixture were carried out with use of an industrial regenerative HRS burner, produced by the Japanese NFK company. The burner, fig. 1, was installed in a semi-industrial chamber. Scheme of the test rig is shown in fig. 2.

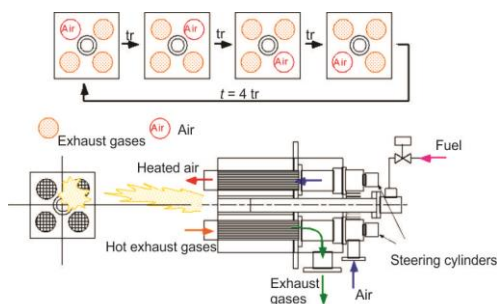


Figure 1. The HRS burner operation scheme

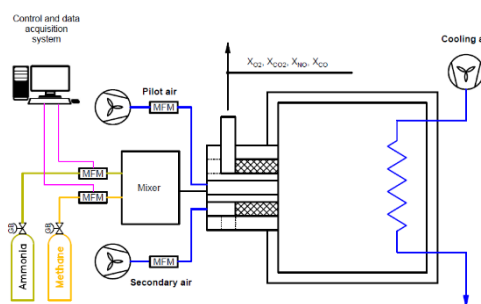


Figure 2. Scheme of the test rig, semi adiabatic combustion chamber

The power firing rate of regenerative burner during the performed tests was 150 kW and resulted in a constant thermal load of the test rig at the level of about 40 kW/m<sup>3</sup>. The HRS burner is equipped with a high-speed fuel nozzle located in the axis of the burner and four air nozzles located on the circumference of the burner. Each of four air nozzles is equipped with a ceramic regenerator placed directly inside the burner housing, giving the possibility of continuous and stable operation of the heat regeneration process. Burner regenerators transferring the heat to the supplied air are made of a collection of ceramic channels called *honeycombs*, with a thermal efficiency of 92%. The operation of the burner is controlled by pneumatic valves providing the cyclic work of air nozzles. Heat regeneration consists of transferring enthalpy of hot exhaust gases to ceramic regenerators and then returning the heat accumulated in the regenerators to the fresh air flowing into the chamber.

To measure the exhaust gases composition, a set of Emerson gas analysers was used. Their main characteristics are presented in tab. 1. The sample of exhaust gases was taken at the outlet chimney of the burner exit, fig. 2. To ensure the process parameters' stability, the power of introduced fuel was adjusted with the use of a mass-flow controller for both methane and NH<sub>3</sub> flow.

**Table 1. Characteristics of the measuring equipment**

Name	Range	Accuracy	Comments
CO <sub>2</sub> (IR detection)	0-25%	±1% of FS	Measurement in the product's exhaust outlet
O <sub>2</sub> (paramagnetic detection)	0-30%		
CO (IR detection)	0-500 ppmv		
NO (chemiluminescence detection)	0-1000 ppmv		
Bronkhorst EL-FLOW F-203AC mass-flow meter	0-400 Nl/min	±0.2% of FS	Measurement of CH <sub>4</sub>
Brooks SLA5850 mass-flow meter	0-25 Nl/min		Measurement of NH <sub>3</sub>

## Methods

Because of the different nature of the NH<sub>3</sub> combustion process in comparison with the methane one (*i.e.* lower laminar flame speed, adiabatic flame temperature, maximum flame temperature, ignition and autoignition temperature, lower calorific value and density, *etc.*), it is necessary to study its impact on the basic parameters in the combustion chamber. The increasing of content NH<sub>3</sub> can also affect the distribution of heat within the combustion chamber, which is crucial in heat transfer processes like steel or glass production. The flue gas recirculation degree, the supplying of regenerated air mode and the amount of oxygen delivered to the chamber can also play a key role in the attaining of a emissive and energetically effective combustion. Therefore, in order to investigate the influence of these factors, it is important to determine the temperature distribution and individual reaction zones by local emissions measurements in the combustion chamber.

During the measurements, the influence of the physicochemical properties of the gaseous fuels and exploitation parameters on pollutant emissions, especially NO and CO, was investigated. The measured values included: the temperature profiles inside the combustion chamber, the molar fraction of pollutant in the exhaust gases, combustion chamber wall temperature and total heat flux (THF).

The research has been performed for parameters of combustion process presented in tab. 2, while the composition of investigated fuels can be found in tab. 3. The main fuel used for the investigation was methane. The NH<sub>3</sub> volume flow rate equal to 2.5, 5.1, and 13.3 N lpm (normal liters per minute) was delivered to the main fuel. It corresponds to 1.0, 2.0, and 5.0% vol. of NH<sub>3</sub> in the fuel stream. Three levels of equivalence ratio (0.75, 0.85, and 0.95) were considered. Additionally, the influence of the regenerators' switching time operation and influence of primary air share on NO emission were studied. The switching time of separate nozzle was ranging from 6 to 12 sec, while the ratio of amount of primary air,  $V_p$ , to

**Table 2. Parameters of the combustion process**

Parameter	Value
The NH <sub>3</sub> content in the fuel [vol.%]	1-5
Main fuel	CH <sub>4</sub>
Fuel bulk velocity [ms <sup>-1</sup> ]	90
Equivalence ratio [-]	0.75-0.95
Temperature of primary air [K]	293
Average temperature of combustion air [K]	1240-1250

**Table 3 Studied fuel composition**

Fuel name	CH <sub>4</sub> [% vol.]	NH <sub>3</sub> [% vol.]
M100	100	0
M99_A1	99	1
M98_A2	98	2
M95_A5	95	5

amount of total combustion air,  $V_{TCA}$ , ranged from 15 to 30%. The burner regeneration unit is built of four honeycomb blocks switching sequentially: one for supplying the air to the chamber and the other

three for rejecting exhaust gases into the exhaust outlet while heating regenerators. The switching time can affect thermal efficiency of the honeycomb blocks [20]. The shorter the air flow time through regenerator, the higher the efficiency of the heat exchange. The calculations performed, describing the efficiency of heat exchange in the regenerators, show that for the analysed range of switching time efficiency of regenerators installed in the HRS burner changes only minimally. For switching time of 6 seconds, the value of  $\eta = 94.4\%$  while increasing this time up to 12 seconds resulted in a 1.5% decrease in heat exchange efficiency of the regenerator. Knowing these results, it was decided to check how switching time affects the emission of NO.

The wall temperature inside the combustion chamber was kept within the range of 1283 and 1320 K. In order to prevent fuel injection velocity disturbances on pollutant emission, this parameter was held at a constant value of 90 m/s.

To account for the dilution of exhaust gases by excess air, the exhaust gases analyser data such as NO and CO concentration are referred to oxygen based reference conditions. For scientific analyses oxygen reference value is equal to 0%. This value was assumed in the calculations. Multiplication factor is calculated and adapted to the obtained values of NO emissions according to:

$$k = \frac{21 - O_{2\text{reference}}}{21 - O_{2\text{measured}}} \quad (1)$$

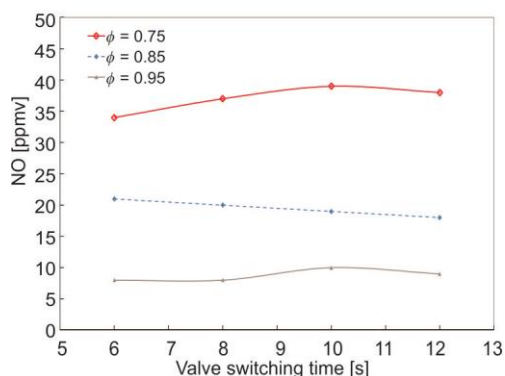
Presented in the paper results of flue gas composition (O<sub>2</sub>, NO, CO) represent the concentration in the dry exhaust gases.

The THF was measured using water-cooled THF probe which consists of three connected to pipes made of austenitic steel. The two outer pipes form the probe jacket filled with cooling water. The internal tube leads wires of thermocouples. The main element of the THF probe construction is the measuring part equipped with thermocouples placed at a known distance in the measuring disc. The probe was calibrated according to Murthy *et al.* [21] standards. The sensor was located on the roof of the combustion chamber, at distances of 500, 1500, and 2100 mm from the burner outlet.

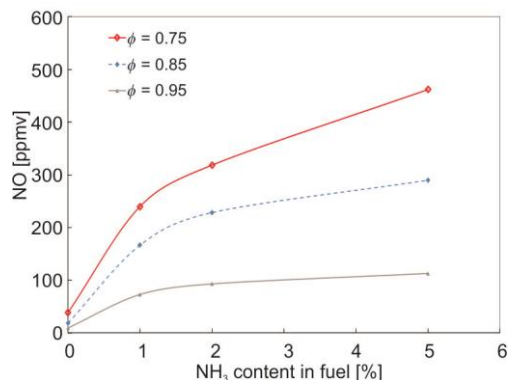
## Results

The results of the influence of the regenerator switching time on NO emissions are presented in fig. 3. It can be noted that the regeneration interval time has a very minor impact on NO emissions in all the investigated cases. A very slight increase, between 6 and 10 seconds, of the regenerator switching time interval was observed for the lowest investigated fuel-air equivalence ratio ( $\phi = 0.75$ ). However, these differences are small for the entire range of the fuel-air equivalence ratio tested. Regarding NO emissions, a very significant effect of fuel-air equivalence ratio in the combustion chamber is present. Increasing of equivalence ratio resulted in strong NO emissions drop, achieving levels below 10 ppmv. It is due to the lack of oxygen needed to create NO from the air nitrogen, according to the thermal mechanism [22, 23].

During the tests with NH<sub>3</sub> the regenerator switching time interval was set to 10 sec. The emission characteristic of CH<sub>4</sub>/NH<sub>3</sub> combustion process is presented in fig. 4.



**Figure 3. Switching time of regenerator influence on NO emissions for M100**



**Figure 4. The NO emissions vs. NH<sub>3</sub> content in the supplied fuel**

Increasing the air amount delivered to the combustion process provides a rise in NO. The intensity of NO formation is most intensive for 0 and 1% vol. of the NH<sub>3</sub> in the supplied fuel for all the investigated fuel-air equivalence ratio levels. However, with increasing the share of NH<sub>3</sub>, this correlation tends to decrease, especially for cases with high fuel-air equivalence ratio ( $\phi = 0.95$ ). In such cases, the increase of NH<sub>3</sub> levels in the fuel has a very slight effect on NO emissions. This is due to the increased conversion of atomic nitrogen to nitrogen (N<sub>2</sub>), as a result of the reaction with NO, it becomes significant in fuel-rich zones. The activity of the  $N + NO \rightarrow N_2 + O$  reaction increases with the increasing levels of NH<sub>3</sub> concentration, resulting in a non-linear NH<sub>3</sub> conversion rate. For cases with  $\phi = 0.85$  and  $\phi = 0.75$ , the influence of NH<sub>3</sub> on NO formation seems to be more significant. An increase of the air amount converts the combustion mixture to be fuel-leaner and the abundance of oxygen intensifies the conversion of NH<sub>3</sub> into NO, which is in agreement with Sullivan *et al.* [18].

The second studied exploitation parameter was the relation between the amount of primary air and the total combustion air  $V_p/V_{TCA}$  delivered to the combustion chamber. The test has been prepared for a constant wall temperature, switching time ( $t = 10$  seconds) and the equivalence ratio equal to 0.85. A positive effect of the increase of primary air share, in case of fuels with NH<sub>3</sub> content, was observed, fig. 5. The NO emissions were reduced by around 15% (reduction by around 48 ppmv for 5% vol. of NH<sub>3</sub>) [19]. The reduction of the combustion air amount by increasing the primary air share causes a reduction of the abundance of oxygen in the main reaction zone and a reduction in NO formation. For methane, where only thermal and prompt NO are formed, the reduction was negligible. The second advantage of increasing the primary air amount is the reduction of carbon monoxide emissions by a factor of more than two for all mixtures of CH<sub>4</sub>/NH<sub>3</sub>, in case of  $V_p/V_{TCA}$ : a change from 15 to 30%. However, it should be noted that the rise of the primary air amount induced decreased the combustion efficiency as more air is delivered to the combustion process without preheating.

In order to determine the emissions of NO formed due to the fuel mechanism; it was necessary to determine a dimensionless conversion factor (CF). This factor determines which compounds containing the molecular nitrogen are converted into NO. The CF is determined according to the eq. (2) where  $n_{NO}$  is the number of moles of NO in the exhaust gases, and  $n_N$

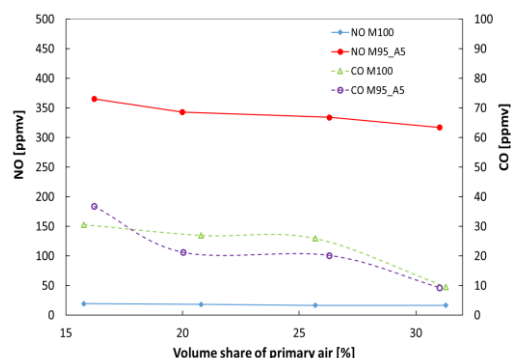


Figure 5. Pollutant emissions vs.  $V_p/V_{TCA}$  ratio

risers, and for  $\phi = 0.75$  it is equal to 8.4% (M95\_A5). Such a tendency was also observed for fuels with 1 and 2 vol.% of NH<sub>3</sub> in the fuel.

is the number of moles of nitrogen compounds (for the case of NH<sub>3</sub>) in the mixture of fuel and air.

$$CF[\%] = 100 \times \frac{n_{NO} [\text{mol}]}{n_N [\text{mol}]} \quad (12)$$

For all the investigated tests with NH<sub>3</sub> configurations, the calculated CF of NH<sub>3</sub> to fuel NO presented extremely low values, tab. 4. The lowest value of  $CF = 2\%$  was obtained for the fuel with NH<sub>3</sub> share equal to 5 vol.% and equivalence ratio  $\phi = 0.95$ . It was noticed that with an increase of the amount of air delivered to the combustion chamber, the value of CF

Table 4. Conversion Factor

Test name	Equivalence ratio $\phi$ [-]		
	0.75	0.85	0.95
	CF [%]		
M99_A1	22.9	15.9	8.4
M98_A2	15.4	10.7	5.9
M95_A5	8.4	5.9	2.0

The most important information describing the combustion process of gaseous fuels in furnace is the temperature distribution. The temperature measurements were carried out at five cross sections of the combustion chamber, located at a distance from the gas burner outlet equal to 150, 650, 1150, 1650, and 2150 mm. Measurements were taken at the height corresponding to the axis of the combustion chamber. The aspiration probe was introduced to the combustion chamber in a horizontal manner. The probe position was controlled by a traverse system with an accuracy of 1 mm. In each cross-section, the temperature was measured at 20 measuring points, distanced 50 and 100 mm, respectively, in the centre and next to the wall of the combustion chamber. The study was prepared for constant operating parameters: switching time  $t = 10$  seconds, equivalence ratio  $\phi = 0.85$  and the average temperature of main combustion air equal to 1220 K. The temperature distribution in the combustion chamber for two investigated fuels (M100 and M95\_A5) are presented in the figs. 6 and 7.

Based on the obtained results, it can be noticed that in the case of fuels with NH<sub>3</sub> content equal to 5%, the maximum measured temperature is slightly lower than for methane. In cases when the burner thermal input is constant, the temperature decrease in combustion chamber for fuel with NH<sub>3</sub> content is caused by a lower maximum combustion temperature of NH<sub>3</sub>. It is also the result of a lower volumetric heat release rate of the CH<sub>4</sub>/NH<sub>3</sub>/air flame compared to the CH<sub>4</sub>/air flame [24]. As the reaction rate of NH<sub>3</sub> is very low compared to methane, the maximum temperature zone could be shifted, relative to pure methane/NH<sub>3</sub> (CH<sub>4</sub>/NH<sub>3</sub>) combustion. Despite this fact, the highest temperature zone is located practically in the same place for both of the investigated cases. To analyse the influence of NH<sub>3</sub> on the temperature distribution field in the combustion chamber, a dimensionless factor,  $R_{tu}$ , proposed by Yang [25] was introduced:

$$R_{tu} = \sqrt{\sum \left( \frac{T_i - \bar{T}}{\bar{T}} \right)^2} \quad (3)$$

where  $T_i$  is temperature measured in a selected point of the combustion chamber and  $\bar{T}$  – the average temperature in the investigated volume.

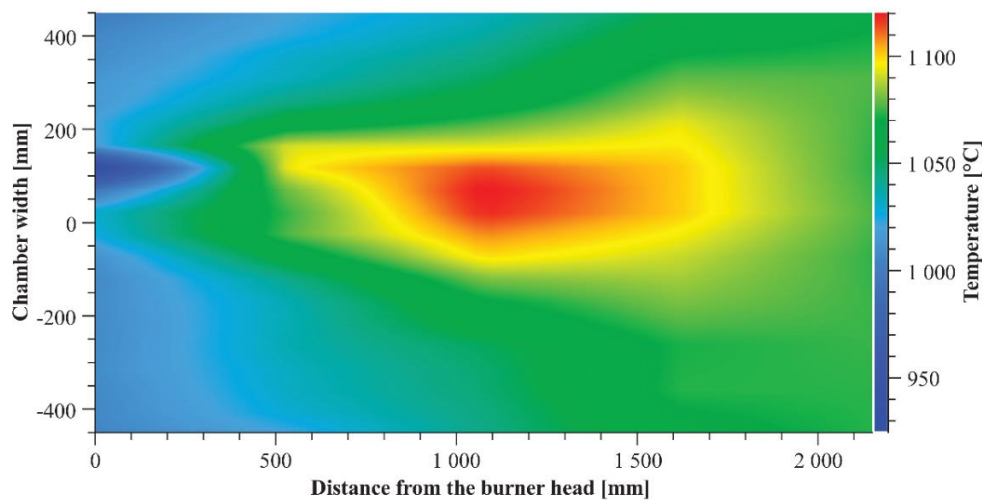


Figure 6. Temperature distribution in the combustion chamber for fuel M100

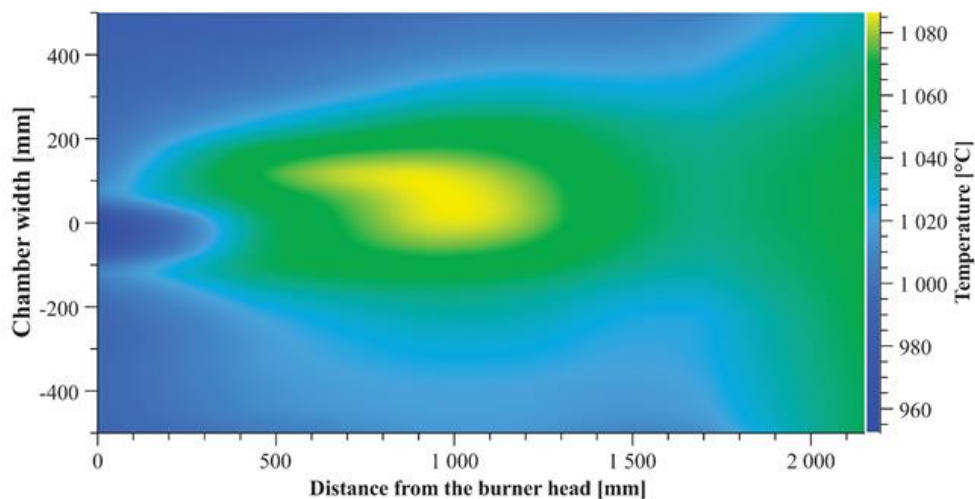


Figure 7. Temperature distribution in the combustion chamber for fuel M95\_A5

The higher the value of the temperature uniformity coefficient, the more diverse the distribution of this value in the studied area. The extreme value of  $R_{tu}$  is the value of a coefficient equal to zero, which corresponds to the flat temperature profile of the gases in the combustion chamber. The average results of  $R_{tu}$  for the analysed cross sections of the combustion chamber are presented in the tab. 5.

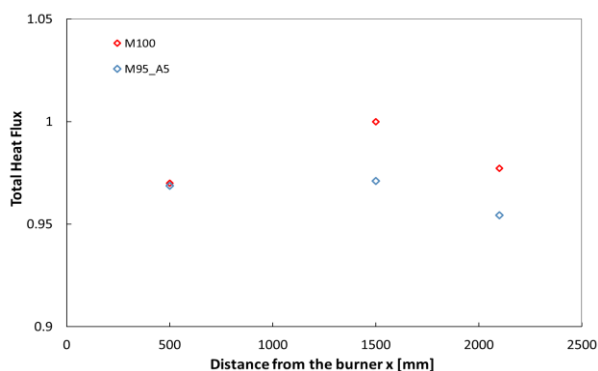


**Table 5. Temperature uniformity  $R_{tu}$  values, depending on distance from the burner plate**

Test name	Distance from the burner head [mm]			
	150	650	1150	1650
	Temperature uniformity coefficient $R_{tu}$ [-]			
M100	0.15	0.10	0.13	0.14
M95_A5	0.05	0.11	0.10	0.07

The results of the temperature uniformity coefficient clearly show that its highest values coincide with the places where the maximum reaction temperature occurs. It can be seen in figs. 6 and 7 that the lowest temperature values are near the walls of the chamber. In contrast, the highest temperature is in the central part of the chamber, at some distance from the fuel nozzle outlets of the burner. Depending on the fuel used, the size of this area varies.

During the studies on the influence of NH<sub>3</sub> content in the fuel on the parameters of flameless combustion technology, the value of the THF was measured. The received value of THF is the sum of the heat delivered to the probe head through radiation and convection. Since in the flameless combustion process the injection velocities (air and fuel) are high, the convection part of heat transfer has a significant impact on THF value. The NH<sub>3</sub> effect on heat transfer from the flame was negligible for the investigated fuels, and the maximum difference was observed for  $x = 1500$  mm. The THF value for this point was reduced by 3%, compared to pure CH<sub>4</sub> fuel. Changes in the THF values in the combustion chamber are shown in fig. 8 presenting data in dimensionless form, where value 1 corresponds to the maximum value of the THF measured for pure methane.



**Figure 8. The THF rate for the investigated fuels, wall temperature  $T_{CH} = 1320$  K, equivalence ratio  $\phi = 0.95$**

## Conclusions

The presented experimental research studies the influence of NH<sub>3</sub> addition into a fuel jet in flameless condition on the operating parameters of an industrial furnace. It was shown that one of the major parameters influencing the NH<sub>3</sub> conversion to NO is the equivalence ratio. Increasing the air availability intensifies the NH<sub>3</sub> to NO conversion. It was found that the conversion efficiency of NH<sub>3</sub> to NO decreases from over 23% ( $\phi = 0.75$ ) at low NH<sub>3</sub>

concentration, to less than 2% at high NH<sub>3</sub> concentration ( $\phi = 0.95$ ). Increasing the NH<sub>3</sub> concentration in the fuel results in a non-linear conversion rate, as consequence of the growing decomposition process of NO to N<sub>2</sub>. The intensity of this process is more visible for richer fuel mixtures. The data analysis has shown that the NH<sub>3</sub> effect on the temperature and the heat transfer distribution in combustion chamber for the investigated volume fractions of NH<sub>3</sub> is negligible. The value of THF decreased by around 3% for CH<sub>4</sub>/NH<sub>3</sub> fuel.

Based on the preliminary investigations, it can be concluded that NH<sub>3</sub>, as an energy storage agent, can be co-fired with methane in flameless combustion technology regime with low pollutant emissions.

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