



## Full Length Article

# Experimental investigation on syngas reburning process in a gaseous fuel firing semi-industrial combustion chamber

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## ABSTRACT

The article presents the results of the experimental investigation of nitric oxides reduction with a reburning process using syngas fuels in semi-industrial gas fired combustion chamber. The studies included the influence of different chemical properties of syngas fuels, on the total of NO and CO emission. The properties of syngas fuels included low calorific value (from 3.43 to 17.2 MJ/Nm<sup>3</sup>), amount of fuel-bound nitrogen and the share of reburning fuel. The tests have been carried out for two main fuels methane and synthesis gas in semi-industrial combustion chamber.

It has been reported that reduction of NO by synthesis reburning fuels reached level up to 80% for low and 45% for medium calorific fuels in case of methane as primary fuel. The high calorific value of synthesis fuel (17.2 MJ/Nm<sup>3</sup>) introduced to the combustion chamber as a reburning agent caused an increase of NO formation especially in case of low primary nitric oxide emission (an increase of 27% – 60 ppmv). The application of reburning process to the Zonal Volumetric Combustion (ZVC) technology allowed to achieve around 43% reduction of nitric oxide emission in case of medium calorific syngas and excess of primary air value equal to 0.5. For the same fuel, with the excess of primary air equal to 0.1 (proffered by ZVC) the decrease of NO was lower, only up to 14%. The highest reduction was observed for low calorific syngas with the highest concentration of ammonia (more than 60% reduction).

## 1. Introduction

The production of heat and electricity in Central and Eastern European Regions is based mainly on coal and lignite. It is the cause of high local emission of air contaminants such as dust, sulfur compounds, nitric oxides carbon monoxide and VOCs. Furthermore, combustion of fossil fuels influences the global emission of carbon dioxide, the concentration of which is increasing and in 2016 CO<sub>2</sub> for the first time remained above 400 ppmv all year [1]. The EU climate and energy policy, introduced the new restriction on greenhouse gases (GHG) emissions such as the “EU 2030” where 40% GHG emission should be reduced by 2030, compared with the 1990 levels or as part of the road map target of 80% emission reduction by 2050. To fulfill these restrictive goals alternative fuels must be considered. There is still a huge amount of potential energy carriers such as biomass, solid municipal waste and landfill gas which can be introduced to the energy sector. Biomass is renewable, environmentally friendly and considered as CO<sub>2</sub> neutral energy source gathers special interest [2]. The share of biomass fuels in Europe in the primary energy consumption is projected to reach

over 15% of EU primary bioenergy supply by 2020 [3]. The share of renewable energy sources in total energy consumption of Poland is expected to rise to 15% by 2020 and 20% by 2030 [4]. It was estimated that waste energy contained only in waste biomass would allow to achieve this goal in many regions of the whole country [5]. The biomass in industry can be used directly as feedstock, for combustion in energetic boilers, co-combusted with coal, as well as converted to liquid biofuels or synthesis gas derived from the gasification process. Direct combustion and co-combustion in energetic boilers cause several problems such as: corrosion (Cl, Na, K), deposition of ash onto boilers heat exchanger and milling difficulties (in case of pulverized boilers) [6]. In paper [7] authors presented the influence of the co-firing of syngas and hard coal on the thermal efficiency of 230 tonnes/h dual fuel pulverized coal-gas fired boiler. The results shows that even 10% share of power delivered in the form of LHV gas (2 MJ/Nm<sup>3</sup>) causes a decrease of boiler efficiency of up to 2% (with syngas share equal to 20% the decrease was up to 4.5%). One of the possibilities to avoid the aforementioned problems is indirect co-firing of pre-processed biomass in the form of syngas or liquid biofuels. Gasification is a very promising technology

*Abbreviations:* CF, conversion factor; COG, coke oven gas; LDA, laser Doppler anemometry; LHV, low heating value; RF, reburning fuel; S<sub>L</sub>, laminar flame speed; T<sub>A</sub>, adiabatic flame temperature; T<sub>SUB</sub>, substrates temperature; ZVC, zonal volumetric combustion; λ<sub>p</sub>, excess ratio of primary air

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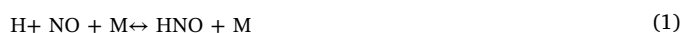
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for energetic purposes, especially in areas where rich resources of biomass are widely available. The synthesis gas consist of flammable components, mainly hydrogen, carbon monoxide and methane, inert gases like nitrogen and carbon dioxide, some amount of unsaturated hydrocarbons ( $C_2H_2$ ,  $C_6H_6$ ) as well as pollutants such as hydrogen sulfide ( $H_2S$ ), ammonia ( $NH_3$ ), tars, halogens and dust. The resulting composition of synthesis gas and its contaminants depends on several factors. These are mainly fuel composition, gasifying agent, process operating conditions (temperature, pressure, residence time) as well as the gasifier type. All of these operating parameters and conditions influence the calorific value of syngas which covers a wide range from 2 to  $19 \text{ MJ/m}^3$ . The synthesis gas obtained from the gasification process has some disadvantages, like low heating value (even below  $2 \text{ MJ/Nm}^3$  [8]), high content of tars (up to 35% for wood gasification [9]) and high content of contaminants ( $H_2S$ ,  $NH_3$ ). For this reason the utilization of synthesis gas as main fuel is difficult because it leads to problems such as corrosion or pipeline blockages due to tar residue. Moreover increased emission of  $NO_x$  (according to fuel mechanism of formation) and  $SO_x$  is observed. Hence the need to develop the existing technologies and conduct investigations of new solutions which can face these obstacles. Flameless combustion is a very promising technology which was successfully implemented in a full industrial system powered by a Copper Blast Furnace [10]. In this installation waste gas with LHV below  $2.5 \text{ MJ/Nm}^3$  is burned in energetic boilers in a semi adiabatic combustion chamber. The principle of Flameless technology, called alternatively HiTAC [11,12], Mild Combustion or Volumetric Combustion is based on separate high velocity injection of gaseous fuels and air to the combustion chamber. It allows to create very strong recirculation zones in combustion chambers. Based on HiTAC principles, very recently, Zonal Volumetric Combustion technology (ZVC) was presented [13]. Different method of indirect utilization of biomass in the form of synthesis gases in existing energetic devices like combustion chambers, combustion pre-chamber of boilers (heat recovery boilers) and boilers themselves is reburning. Reburning is a method of nitric oxides reduction, whose efficiency is determined by reactions taking place in the so-called rich zone. Reactions between hydrocarbons radicals and nitric oxides creating HCN (hydrogen cyanide) are most responsible for this positive effect. HCN converts to  $NH_i$  species ( $i = 0$  to  $i = 3$ ) with participation of hydrocarbon radicals and hydroxyl. Finally  $N_2$  is created by reactions of  $NH_i$  with  $NO$  [14]. Mechanism of the  $NO$  reduction by hydrocarbons reburning process is presented in Fig. 1. The rate of these reactions depends on composition of the reburning zone (concentration of nitric oxide and  $O$ ,  $OH$ ,  $H$ ,  $CH_i$  radicals). The efficiency of reburning for hydrocarbon fuels ranges from 30 up to 70% depending on operation parameters.

The hydrocarbons and solid fuels as reburning agents have been extensively investigated in the past and mechanism of nitric oxides

reduction by them is well recognized [7,16–19]. Maly et al. [20] investigated advance reburning approach for different types of reburning fuels and achieved even 96% of  $NO_x$  reduction for biomass, 78% for coal fines and 91% for natural gas. In [21] Ballester et al. investigated natural gas and biomass effect on reburning and achieved  $NO_x$  emission reduction by 62.7 and 62.8% respectively.

For flammable components of syngas fuels such as  $CO$  and  $H_2$  the ability of  $NO_x$  reduction is lower. The calculated  $NO_x$  reduction efficiency by non-hydrocarbon components was in the range of 20–30% and increased slightly with the increase in temperature and fuel/air ratio in the reburning zone [22]. Better understatement this phenomena requires taking into account a group of possible chemical reactions undergoing. Glarborg et al. [23] studied the  $NO$  chemistry in the absence of hydrocarbon radicals and reactive nitrogen species. They stated that in presence of  $O_2$  the  $NO$  removal potentially occurs through the reaction sequence:



followed by reaction of  $NH$  with  $NO$  and  $OH$ , resulting in  $N_2$  and  $HNO$  formation respectively. In the absence of oxygen,  $NO$  is removed at medium temperatures by the relatively slow sequence:



In the presence of  $CO$  a significant fraction of  $NO$  could be removed by reactions with  $CO$  at higher temperatures, but reactions appear to be slow up to  $1140 \text{ K}$ , as well as reaction of  $NH$  with  $H_2O$ . Presented reactions could justify correctness of above-mentioned thesis, although exact identification of the chemical paths in the flow conditions involves enhanced numerical study on described process. Numerical calculations could state as logical following step for the ZVC reburning research.

Werle [24] presented the numerical results of  $NO_x$  reduction process during low calorific syngas combustion in the coal fired boiler. The maximum  $NO_x$  reduction factor was received for about 15% share of energy delivered with fuel reburning. Wilk et al. in [25] presented the numerical calculations of  $NO_x$  reduction process using sewage sludge delivered syngas in a natural gas fired reactor. It was evaluated that for a syngas share of up to 15%  $NO_x$  emission level decreased ( $NO_x$  reduction efficiency was up to 44%), while addition of 19% of syngas lowered the  $NO_x$  reduction efficiency to 21%. In paper [26] Xiaoying Hu and co-authors presented the effect of syngas reburning process on the emission of  $N_2O$  and  $NO$  in a Circulating Fluidized Bed boiler (CFB). For each studied case they received  $N_2O$  reduction rate in the range from 22 to 99% and  $NO_x$  reduction factor from 35 to 54%.

Next to non-hydrocarbons components the synthesis gases received from gasification of biomass contains tars, which can improve the efficiency of reburning process. Tars consist of complex mixtures of organic compounds such as aromatic and heteroaromatic species as well as polycyclic aromatic compounds (PAHs). The main components are phenol, benzene, toluene, naphthalene and pyridine [9]. In paper [27] authors presented experimental and numerical investigation of  $NO_x$  reduction by syngas reburning, where phenol was a reference component of tar. The experimental reduction efficiency was around 85%.

As aforementioned ZVC allows to significantly reduce  $NO$  emission [13]. Therefore it has been decided to extend research of this technology on investigations of reburning effect with synthesis gases as reburning fuel. The main objective of the present experimental study was evaluation of nitric oxides reduction ability in semi-industrial gas firing combustion chamber (methane or syngas) by low, medium and high calorific value syngas as a reburning agent.

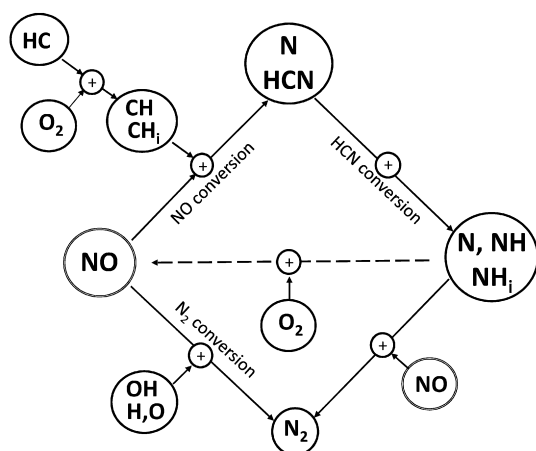


Fig. 1. Mechanism of  $NO$  reduction by hydrocarbons reburning [15].

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