

NO_x Formation by Synthesis Gas – Natural Gas Co-firing

K. Valler^{1*}, Á. Wopera¹, Á. B. Palotás¹, K. J. Whitty²

¹Department of Combustion Technology and Thermal Energy, University of Miskolc, Hungary

²Department of Chemical Engineering, University of Utah, USA

Abstract

Synthesis gas from biomass gasification was examined by co-firing with natural gas. Calculations were performed to study the effect of increasing the fraction of synthesis gas in the syngas- natural gas fuel mixture on flame temperature and other combustion parameters. Combustion of this mixture was achieved in two different furnace and by two different firing method. NO_x and CO₂ emissions have been measured and compared. The combined utilization of a biofuel –derived syngas and natural gas was found to be a realistic option for the use of renewable energy in existing gas-fired fossil fuel systems.

Introduction

The process, called gasification produces synthesis gas through the chemical conversion of biomass, usually involving partial oxidation of the feedstock in a reducing atmosphere in the presence of air, oxygen and/or steam. The gasification agent allows the feedstock to be quickly converted into gas by means of different heterogeneous reactions [1]. Syngas consists primarily of hydrogen, carbon monoxide, and very often some carbon dioxide, and has less than half the energy density of natural gas. Syngas is combustible and often used as a fuel source or as an intermediate for the production of other chemicals [2].

Gasification process offers a number of important advantages over the more conventional combustion process: gas has much better burning properties than solids, it is easier to control and it produces less particulate emissions and gaseous pollutants [1].

Beside the synthesis gas landfill gas (LFG) was also examined. Landfill gas results from anaerobic decomposition of the biodegradable organic materials contained in municipal solid wastes (such as food waste, paper, textile, wood, etc.), is a flammable and potentially harmful gaseous mixture consisting mostly of methane (CH₄) and CO₂ together with a number of volatile organic compounds (VOC) [3].

The aim of the study was to examine how the above detailed synthesis gas and landfill gas can be combusted with natural gas and what the effects are on forming NO_x.

Reburning, or staged fuel injection that we applied is a known combustion technique for reducing NO_x emissions. In reburning, fuel is injected into the combustion products downstream of the combustion zone. Literature in the area of reburning is related to the subject of NO_x minimization from by-product fuel combustion in that these techniques can be associated with lower peak flame temperatures in the primary combustion zone, as some of the primary fuel is used as the reburn fuel [4-6].

Introduction of the furnace and the firing method

The experiment introduced in this study was accomplished in a laboratory test furnace at the University of Miskolc. These results were compared to those obtained from the research carried out by a different firing method at the University of Utah.

Co-firing of natural gas and different bio- derived syngases in a distributed fuel injection scheme have been studied for several years at the Department of Combustion Technology of the University of Miskolc. The experimental scheme that has been utilized involves the supply of the total combustion fuel in two stages within the burner, with air being supplied only in the first stage.

The laboratory test furnace that was used for the experiments has an air cooling system to control the furnace temperature. Experiments were carried out at a constant firing rate of 20 kW.

Operation must be carried out under fuel lean condition to minimize CO production while not significantly increasing NO emissions. That's why stoichiometric ratio (SR) of 1.1 was used. Figure 1 shows the two-stage INOX burner designed and produced for experimental purposes. It can be operated both in a traditional single stage operational mode and with two-stage firing. During these experiments, the syngas was injected in the second stage.

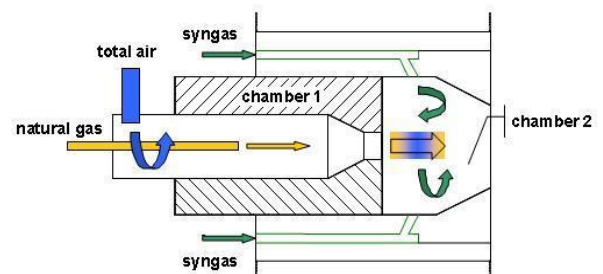


Figure 1: Test burner injection locations

* Corresponding author: vallerk@gmail.com

Associated Web site: <http://combustion.uni-miskolc.hu>

European Combustion Meeting 2009

Two different sort of synthesis gases used in some former experiments in the University of Miskolc are presented in this study. These are named as 'syngas 2' and 'syngas 3' for distinction from the other gases examined. These synthesis gases were provided as preprepared mixtures from gas tanks.

The composition of syngas 2 was 10% CO₂, 5% CH₄, 50% N₂, 25% CO, 10% H₂ with a heating value of 6,027 kJ/m³. This synthesis gas resembles the producer gas from air gasification due to its high (60%) inert content.

The other synthesis gas utilized (syngas 3) contained 35,8% CO₂, 21,2% CH₄, 19,7% CO, 23,3% H₂ with a 12,600 kJ/m³ heating value. This composition is similar to gas generated from oxygen gasification or pyrolytic gasification.

The other furnace that was used for the experiments is located at the University of Utah. This furnace is actually a downwards flowing natural gas-fired afterburner belonging to the University of Utah gasification test system.

The afterburner (Figure 2) is a 4,6 meter tall vessel lined with two layers of refractory to make a 0,35 m diameter reaction chamber. Gas enters the top of the vessel and is combusted by two opposing natural gas burners in the top two ports. The gas is burned at a minimum temperature of 1093°C.

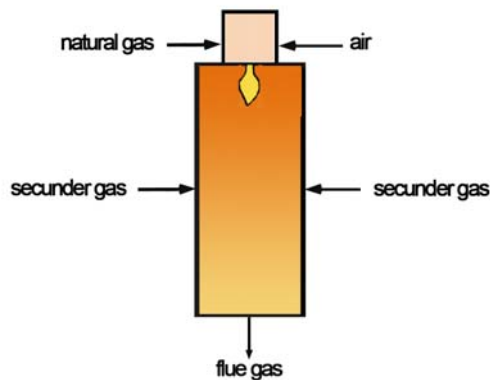


Figure 2: Schematic diagram of the University of Utah afterburner

Additional ports downstream are provided for sampling or viewing, and two lower ports were used also to feed the synthesis gas to the burner in this experiment. The afterburner is operated under slightly negative pressure, provided by the facility's indirect-draft fan.

In fuel reburning the synthesis gas as a secondary fuel source was introduced downstream of the secondary combustion zone. Experiments were carried out at a constant firing rate of 30 kW and stoichiometric ratio of 1.1.

The natural gas used in the study was obtained from the local utility company.

The synthesis gases and landfill gas used in this experimentation were simulated dry gas mixtures, prepared from individual cylinders of H₂, CO, CO₂, CH₄ and N₂.

The compositions of the synthesis gases were the same as those used in the University of Miskolc experiments.

Landfill gas contains typically about 50% methane and 50% carbon-dioxide, that is why we used the same composition. Heating value of it was 17,898 kJ/m³. In fact landfill gas also contains less than 1% sulfides and non-methane organic compounds, however these impurities were not considered in this investigation.

Calculations

First, calculations were performed to study the effect of increasing the fraction of the secondary gas in the syngas – natural gas or landfill gas – natural gas fuel mixture on flame temperature and other combustion parameters.

Because of the strong relation between adiabatic flame temperature (AFT) and NO formation the change of the AFT were calculated in the function of the secondary gas fraction.

Figure 3 shows the change in adiabatic flame temperature as the fraction of secondary gas (syngas, LFG) is increased.

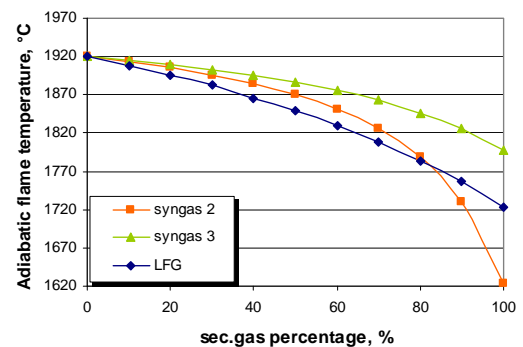


Figure 3: Change of adiabatic flame temperature at different percentages of syngas/landfill gas-natural gas

According to the calculations, the adiabatic flame temperature drops by increasing the % of syngas rate in the mixture due to its lower heating value as compared to natural gas.

Figure 4 shows the change in the Wobbe index as a function of gas mixture. It can be a complex task to burn some of the mixtures, in particular at larger rates of syngas, because of the necessary modifications to the burner.

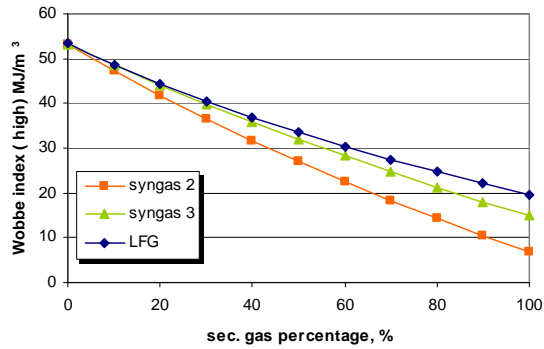


Figure 4: The change of high Wobbe index at different percentages of syngases and landfill gas

Results and discussion

Figures 5 and 6 summarize the results for the measurements of NO_x reduction and CO₂ formation with different natural gas – synthesis gas mixtures examined in the Miskolc furnace.

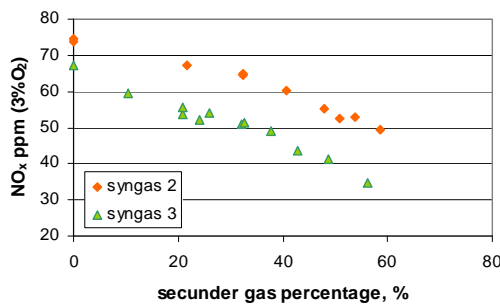


Figure 5: Change of NO_x formation at different percentages of primary and secondary gas (mixture: natural gas – syngas2,3) (Firing rate=20 kW; T_{max}=1050 °C; SR=1,1)

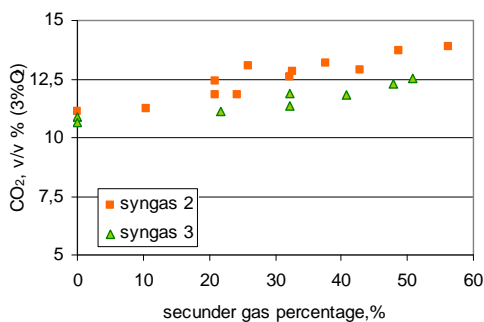


Figure 6: Change of CO₂ formation at different percentages of primary and secondary gas (mixture: natural gas – syngas2,3) (Firing rate=30 kW; T_{max}=1050 °C; SR=1,1)

From the above charts (Figures 5-6) NO_x reduction can be seen while increasing the amount of secondary gas in the natural gas – syngas mixture. At the same time CO₂ level is rising.

Figure 7 shows the results of NO_x and CO₂ emission from landfill reburning. Natural gas was fed into the primary combustion chamber for normal firing (0 sec. gas below), and increasing amounts of secondary gas were injected in the second stage, while maintaining a constant overall firing rate. These results were compared to the data of staged firing with increasing levels of secondary gas. Experimental results were corrected to 3% O₂ content in the flue gas. Figure 7 illustrates a decrease in NO_x emissions with an increase in fuel staging for landfill gas. This chart also shows a slight increase in CO₂ level.

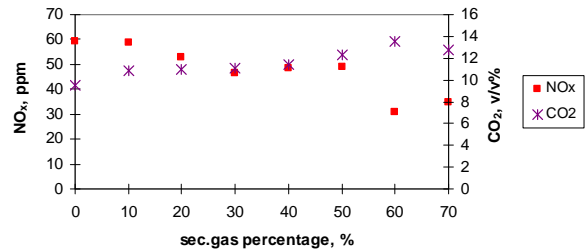


Figure 7: Change of NO_x formation at different percentages of primary and secondary gas (mixture: natural gas – landfill gas) (Firing rate=30 kW; T_{max}=1050 °C; SR=1,1)

On Figure 8 the rate of NO_x reduction can be seen for each measurement by the two different furnaces.

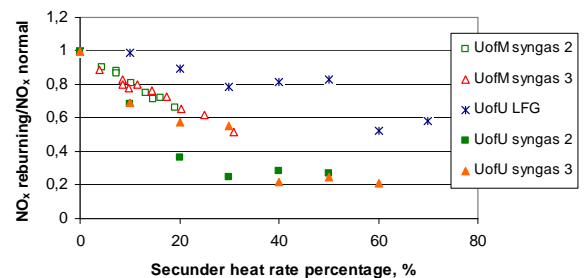


Figure 8 : NO_x reduction at different rate of secondary and total heat flow (natural gas – LFG/ syngas 2-3); (Firing rate 20 kW for University of Miskolc and 30 kW for University of Utah furnace; T_{max}=1050 °C; SR=1,1)

The illustration shows clearly that increasing the amount of secondary gas in the fuel blend, the NO_x formation will decrease. The degree of the reduction is similar for each syngas with no regard to the furnace. Landfill gas mixing performed the less NO_x reduction. Both syngases are seemed to be effective as a reburning fuel.

Figure 9 represents the NO_x reduction as a function of SR in the primary combustion chamber, which has a specific part in the NO_x formation. The results of staged firing of pure natural gas measured in the Miskolc furnace are also included in this diagram.

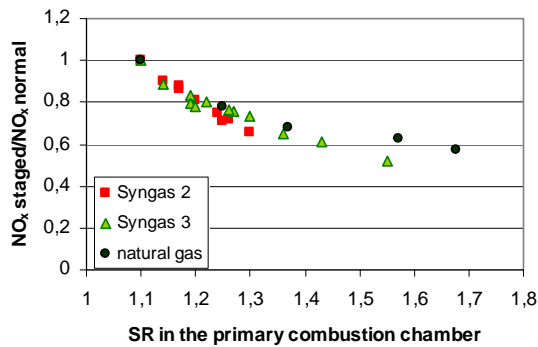


Figure 9: NO_x reduction as a function of the SR of the primary combustion chamber (Firing rate =20 kW; $T_{\text{max}}=1050\text{ }^\circ\text{C}$; SR=1,1)

The illustration shows, that increasing the amount of air in the primary combustion chamber will reduce the NO_x formation due to the flame temperature drop.

Conclusions

This paper presents a potential application of biomass energy sources to replace some part of fossil fuel combustion. The potential use of fuel reburning as a means of reducing nitrogen oxide emissions from the combustion of natural gas was examined.

Gasification of biomass provides a possibility to get a good quality syngas having excellent firing and emissions properties (low NO_x formation and particles). The mixing of syngas and landfill gas with natural gas influences the adiabatic flame temperature as well as the firing parameters. Therefore the combustion of syngas is more advantageous as a secondary gas (through a dedicated pipeline), especially for those with significantly different Wobbe indices.

Since a secondary gas with high CO_2 content is used to replace natural gas, higher CO_2 emission may occur in the flue gas. However it doesn't contribute to the greenhouse effect when the secondary gas comes from a renewable source.

Although the similarity of the NO_x emission results from each furnaces wasn't expected because of the different furnace design and operating heat rate, the reduction tendency can be seen clearly.

There is strong relationship between stoichiometric ratio of air supply and NO_x emission. Natural gas blending with lower heating value gases is reducing NO_x emissions as a result of thermal dilution.

The combined utilization of syngas and natural gas are realistic options for use of renewable energy of the existing fossil fuel system.

Acknowledgements

This research has been founded by the Hungarian National Research Foundation (OTKA T 029199 and T 046471) and the Cooperation Research Center in Mechatronics and Materials Science at the University of Miskolc.

References

- [1] Downdraft/ Updraft gasifier for syngas production from solid waste <http://www.freepatentsonline.com/WO2007081296.htm>
- [2] Beychok, M.R., Coal gasification and the Phenosolvan process, American Chemical Society 168th National Meeting, Atlantic City, September 1974
- [3] Xiaoli Hao, Hongxing Yang, Guoqiang Zhang : Trigeneration: A new way for landfill gas utilization and its feasibility in Hong Kong, Energy Policy, Vol.36., 2008. October
- [4] Shoffstall, D.R. (1977) Burner design Criteria for NO_x Control from Low-Btu- Gas Combustion: Volume I. Ambient Fuel Temperature. Institute of Gas Technology, U.S. Environmental Protection Agency, EPA-600/7-77-094a
- [5] Waibel, R.T. and Fleming, E.S. (1979) Development of Combustion Data to Utilize Low-Btu Gases as Industrial Fuels. Institute of Gas Technology, U.S. Environmental Protection Agency, EPA- 600/7-78-191
- [6] Shoffstall, D.R., and Waibel, R.T. (1977) Burner design Criteria for NO_x Control from Low-Btu- Gas Combustion: Volume II. Elevated Fuel Temperature. Institute of Gas Technology, U.S. Environmental Protection Agency, EPA-600/7-77-094b