CHEMICAL ENGINEERING TRANSACTIONS

VOL. 37, 2014

Guest Editors: Eliseo Ranzi, Katharina Kohse- Höinghaus
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**ISBN** 978-88-95608-28-0; **ISSN** 2283-9216

A publication of
AIDIC
The Italian Association
of Chemical Engineering
www.aidic.it/cet

DOI: 10.3303/CET1437003

# Investigations at a Micro-Scale Installation Regarding Emission Reduction by Air Staging and Integrated Catalysis

Ingo Hartmann, Mario König, Mirjam Matthes*

DBFZ Deutsches Biomasseforschungszentrum gemeinnützige GmbH, Thermo-chemical Conversion Department,
Torgauer Straße 116, D-04347 Leipzig, Germany
Mirjam.Matthes@dbfz.de

The use of biomass for heat production in small scale combustion systems provides an on-site energy supply from renewable resources. Low pollutant emissions as well as heat output on demand are the basic requirements for the application of environmentally friendly and efficient systems.

Investigations regarding pollutant formation have been carried out at a micro-scale combustion system with a heat output between 1 and 2 kW. The flue gas composition under different combustion conditions was observed by variation of primary and secondary air supply. In this manner, the combustion with excess and deficient air was investigated amongst others. Beside the occurrence of incomplete oxidized carbon compounds (carbon monoxide, volatile organic compounds) also the influence of the air supply on nitrogen oxide concentration was monitored. The installation allows the integration of catalysts in different temperature zones. Preliminary considerations regarding the operation of the micro-scale system with a catalyst have been carried out.

## 1. Introduction

In line with improved thermal insulation of buildings heat production furnaces in the low power range gain relevance. Furthermore, the use of renewable resources is promoted to reduce the release of greenhouse gas from fossil fuels to the atmosphere. The formation of pollutants is not completely avoidable during the operation of small-scale biomass combustion systems. A complete conversion of the combustion gas to carbon dioxide and water can be promoted by integration of catalysts. Challenges regarding low emission operation and the use of catalysts in combustion systems are the unsteady conditions and the complex gas composition. Deficient or excess air as well as fuel overload occurs during part load and incorrect operation phases. In addition, the combustion of non-wood biomass in small-scale systems causes often higher emissions (Carroll and Finnan, 2013). The performed investigations refer to the two mentioned key aspects: Pollutant formation in small-scale biomass combustion systems and the possible emission reduction by integration of catalysts. Micro-scale combustion systems allow fundamental research of combustion processes. Investigations with laboratory devices describe that the conversion process of biomass and the flue gas composition is dependent on several conditions like temperature and residence time in the gasification and combustion zone (Pettersson et al., 2010), fuel composition (Brunner et al., 2013) as well as air supply (Skreiberg et al., 2012). For example, the amount of chlorides and sulfates in the fuel has an impact on particulate matter concentration in the flue gas (Sippula et al., 2008). The purpose of the constructed micro-scale system used for the experiments presented in the following sections is slightly different from the subject matters mentioned in the cited articles. The construction of an automatically fed downdraught biomass combustion system with a low nominal heat output and the achievement of low emission operation is the focus of the investigations. In this context, primary emission reduction measures and secondary measures integrated in the installation are of interest. The selection of a downdraught combustion system with staged air supply can provide a high conversion rate and low particulate matter emissions. It is already widespread for wood-fired boilers (Kinsey et al., 2012) and also applied for stoves (Specht and Specht, 2003).Flue or rather process gas cleaning with respect to carbon monoxide and volatile organic compounds (VOC) is already a known measure in process technology, i.e.

Please cite this article as: Hartmann I., Koenig M., Matthes M., 2014, Investigations at a micro-scale installation regarding emission reduction by air staging and integrated catalysis , Chemical Engineering Transactions, 37, 13-18 DOI: 10.3303/CET1437003

for treatment of waste gases (Jecha et al., 2013). The catalytic oxidation of CO and VOC can be carried out with noble metals and transition metal oxides (Heck et al. 2009). Either the use of a porous support material or their manufacturing in a porous solid form is necessary to gain a low pressure drop and accordingly small impact on the pressure in the combustion system. Various options of porous systems are applicable from honeycombs or wire meshes (Heraeus, 2013), packed beds to foams out of metal (Alantum, 2013) or ceramic materials each with different cell densities or rather porosities.

## 2. Experiments

### 2.1 Experimental setup of micro-scale installation

The constructed micro-scale installation is a combustion system with a heat output between 1 and 2 kW. Its design was developed during several tests aiming to the implementation of a continuously operated low power biomass firing system (König et al., 2013). The conversion is designed as a two-stage process with gasification and combustion according to the downdraught principle. Comminuted wood pellets have been used as fuel and are supplied with a screw feeder from a fuel tank to ensure continuous operation. The supply is adjustable by variation of speed frequency of the screw feeder. The fuel is fed in a stainless steel tube and falls on a grate, which allows the passage of gas with low pressure drop and reduces the passage of unburned fuel particles. The air is supplied in two stages analogous to the conversion process. Primary air is provided above the grate and secondary air below the grate. Defined volume flows of primary and secondary air are introduced with the use of mass flow controllers. The gas passes from the stainless steel tube into a silica glass reactor serving as combustion chamber. There, flaming combustion takes place. Downstream of the glass reactor, a heat exchanger block is attached. Finally, before the chimney fan a measuring section for flue gas characterization is available. The stainless steel tube is enclosed by a tube furnace for heating of the grate zone and realization of the ignition procedure. During combustion arising ash particles are gathered at the bottom of the installation inside the heat exchanger block and have to be removed periodically.

### 2.2 Combustion operation

The fuel used in the experiments was prepared from wood pellets (EN plus certified) with a calorific value of 19.1 kJ/kg dry matter basis. Previous feed tests with sifted and not sifted samples comminuted to different fuel particle sizes showed the highest and most homogeneous dosage with not sifted samples (König et al., 2013). The pellets have been comminuted to a size of 6 mm. Fuel supply was adjusted to 0.33 kg/h during ignition procedure and 0.27 kg/h during continuous operation. This results in a power level of 1.75 kW and 1.43 kW respectively. Primary and Secondary air was varied during continuous operation to investigate different conversion conditions. Analogous to air regulation procedures of boilers and stoves, the ignition procedure was performed with a high primary air rate to heat up the fuel bed at the grate by inducing high conversion rates and accordingly a temperature increase. The tube furnace had a heating temperature of 825 K throughout the whole experiment. Continuous operation was started with the reduction of primary air rate and fuel supply rate, when a gas temperature of 1000-1100 K was reached below the grate. The temperatures in the silica class reactor respectively the combustion chamber have been between 550 and 1150 K, with the maximum about 10 cm below the passage from the stainless steel tube with the grate into the silica glass reactor.

### 2.3 Measurement methods

The gaseous flue gas composition was continuously analyzed in the measuring section after the heat exchanger block. The amount of carbon monoxide (CO), nitrogen oxides ($NO_X$), carbon dioxide ($CO_2$) and several hydrocarbons for example methane were measured by Fourier transform infrared spectroscopy. A quantification of the gas concentration was carried out for the first three compounds. For the hydrocarbons a qualitative analysis has been done. The oxygen concentration was determined with a paramagnetic analyzer and the total concentration of volatile organic compounds (VOC) with a flame ionization detector. For two air supply settings having the lowest concentration of unburned carbon compounds (CO and VOC) a gravimetric dust measurement was carried out. An isokinetic measurement was not possible with the given setup of the test stand. The received data is only considered as first indication value for this reason. Modification of the measuring section is intended for further investigations. The temperature profile in the silica glass tube was recorded with seven thermocouples from the nozzle below the grate to the heat exchanger block inlet. In addition, temperature was measured in the measuring section as well as the static pressure. The static pressure in the installation was held constant by a chimney fan regulation.

Operation of the installation with respect to the combustion process has been analyzed for different conditions by adjustment of primary and secondary air supply. Average values for the flue gas composition have been determined for continuous operation over time intervals of at least 25 min. Only the data of stable state flue gas composition was used.

## 3. Results and discussion

### 3.1 Influence of air supply on flue gas composition

The ignition procedure carried out during the experiments is one approach for starting the operation of the installation. The performed investigations clearly demonstrated the drawbacks of this procedure. Inadequate heat transfer and long heating time is given at the present development state. The concentration of unburned compounds in the flue gas during ignition is higher than the measurement range of the used analyzers. For this reason, further developments with respect to more effective ignition procedures are carried out.

At continuous operation high temperatures up to about 1100 K and low emissions have been achieved. The combustion process has been analyzed with different primary and secondary air supply rates. The objective target was the observation of different conversion states including optimal operation as well as deficient air and excess air. Repeated testing has been carried out for some air settings to determine reproducibility of the results. In Figure 1 the results for the measurement of oxygen and CO concentration for four different air supply settings are presented. A variation of the concentration values is evident.

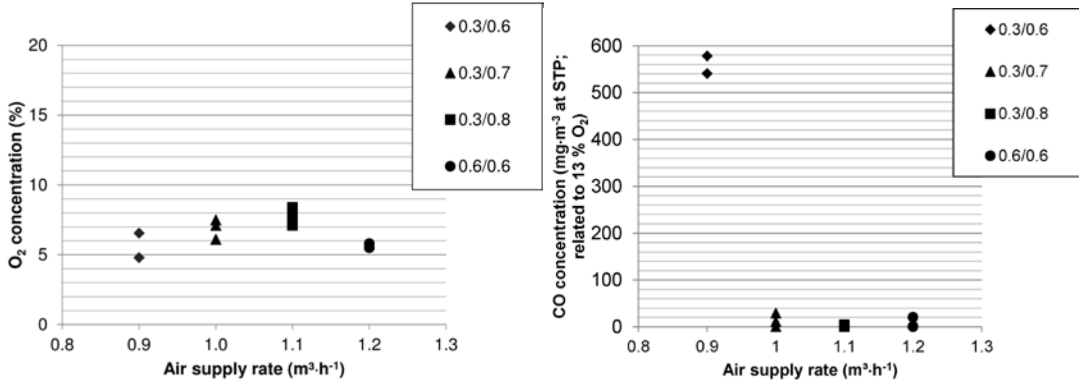

*Figure 1: Oxygen concentration (left figure) and carbon monoxide concentration (right figure) in the flue gas for combustion operation with four different air supply settings (indication: primary/secondary air volume flow in $m^3 \cdot h^{-1}$)*

There are three main reasons for these variations: measurement error of the analyzers, inhomogeneity of biomass combustion and air leakage in the installation or measuring section. Biomass composition and measurement error of the analyzer are given specifications. According to a balance study for the conversion process of the biomass with air considering fuel composition, air supply rate and measured flue gas composition, it is assumed that the measurement results are influenced by a relevant amount of inleaked air. Based on the calculation the ratio the inleaked air accounts 30 to 40 % of the total air volume dependent on operation mode. Since normalized flue gas composition related to 13 % oxygen shows adequate variance of the results for biomass combustion systems, the leak position is supposed to be after the reaction zone before or at the measurement section. The construction of the installation and measurement section has to be advanced with respect to tightness of the whole system to gather more precise results, especially for oxygen values.

Nevertheless, important aspects regarding the influence of air supply on combustion performance could be observed with the current application. In Figure 2 and Figure 3 CO and $NO_X$ concentration for different air supply settings are diagramed combined with the corresponding oxygen concentration. The operation with concentration of CO and VOC below 10 mg/m³ (related to 13 % $O_2$; STP) was possible with two different primary and secondary air supply rates. A primary air rate of 0.3 m³/h and a secondary air rate of 0.8 m³/h resulted in low level of unburned carbon compounds in the flue gas. The flame appeared below the secondary air inlet and had a yellowish color. The second operation mode for low CO and VOC concentrations in the flue gas was achieved with 0.6 m³/h volume flow rate for both primary and secondary air. A red-blue flame appearance was observed below the nozzle for this operation mode. On the basis of these two operation modes two different conversion options can be considered. Since the yellow appearance of flames is related to soot particle formation, it may be assumed that for the first version a lower conversion state is achieved in the primary zone at the grate and then total oxidation of higher molecular burnable organic components took place in the secondary zone. For the second setting with higher primary air rate a higher conversion rate at the grate can be assumed and therefore the flame appearance may be dominated by excited hydrocarbon radicals with one or two carbon atoms in the radical molecule (C1- and C2-radicals). Two relevant processes are described in literature for the formation of flames. On one hand combustible gases and oxygen can be premixed and be heated until ignition temperature is achieved at the flame front, on the other hand diffusion flames are formed when no sufficient oxidizer is present in the fuel and oxygen is supplied from the surrounding by diffusion. Since the air supply in the primary or rather gasification zone is quite higher for the second operation mode with 0.6 m³/h primary air, the different flame appearance may be also interpreted by assuming a higher premixing rate and a lower diffusion impact on flame formation. Apart from the results observed for the CO and VOC concentration, a significant difference was found for the nitrogen oxides concentration. During operation with the higher primary air rate and lower secondary air (0.6/0.6 m³/h) the lower $NO_X$ concentration was measured.

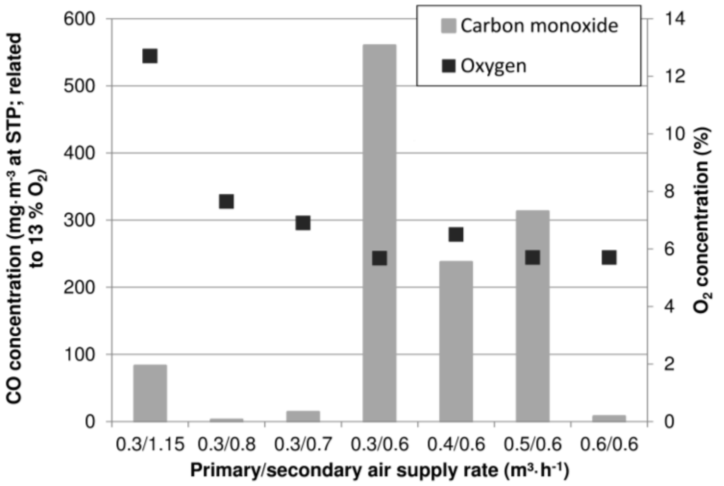

*Figure 2: Carbon monoxide and oxygen concentration in the flue gas for combustion operation with different air supply settings*

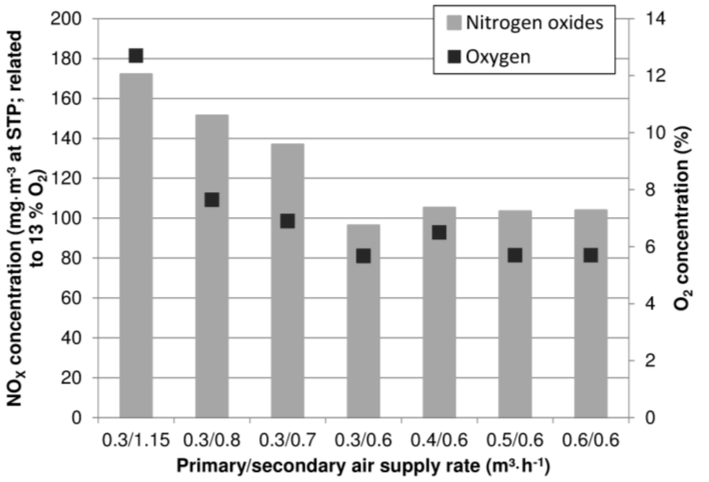

*Figure 3: Nitrogen oxides and oxygen concentration in the flue gas for combustion operation with different air supply settings*

The comparison of all measuring data determined during the experiments showed a dependency of the $NO_X$ concentration from secondary air and residual oxygen respectively. The variation of primary air resulted only in minor changes of $NO_X$ concentration. Results for $NO_X$ concentration in relation to primary and secondary air rate are shown in Figure 4. Skreiberg et al. (2012) have also investigated the dependency of $NO_X$ formation to the air ratio using a small-scale installation with upper combustion and staged air supply. They found an optimum primary air ratio under substoichiometric conditions in the primary zone with an excess air ratio of about 0.75. This is consistent with the here described results. Though, contrary to Skreiberg et al. (2012) a significant correlation of secondary air and not primary air was observed with the given data. Therefore, further investigations are of high interest. For both air supply settings the gravimetric measured particulate matter concentration was below 5 mg/m$^3$ (related to 13 % $O_2$, STP). Deficient air had a higher impact on the concentration of CO and VOC than excess air in the experiments. The reduction of secondary air by 0.2 m$^3$/h starting from the air setting of 0.3/0.8 m$^3$/h for primary/secondary air rate led to high concentrations of CO and VOC in the flue gas. The increase of secondary air by 0.35 m$^3$/h resulted only in a comparably lower increase of CO concentration and no change in VOC concentration.

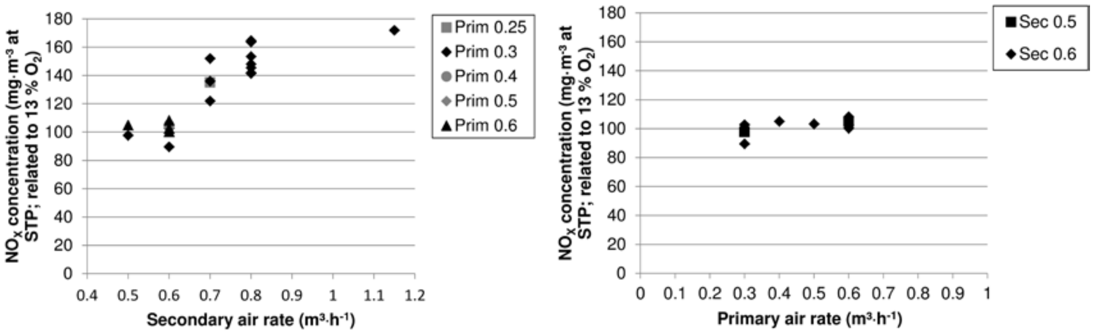

*Figure 4: $NO_X$ concentration in relation to the secondary (left figure) and primary (right figure) air rate (indication: corresponding primary air rates for the left graph and secondary air rates for the right graph respectively in $m^3 \bullet h^{-1}$)*

### 3.2 Considerations regarding emission reduction by catalysis

The variation of secondary air in the experiments showed the need of emission reduction during combustion phases with deficient and excess air. A catalyst can promote the total oxidation of the measured pollutants CO and VOC. Beside the modification of the reaction process due to the catalytic surface, changes of temperature, static pressure and fluid dynamics occur. Therefore, investigations regarding the change in process parameter caused by the integration of a monolith have to be carried out.

Ceramic foams offer advantageous qualities as catalyst support. Their application in flue gas cleaning is promising because of their abilities with respect to radial gas mixing, heat and mass transfer as well as pressure drop (Patcas et al., 2007). The use of catalytic active materials supported on alumina is widely used for catalytic applications (Heck et al., 2009). Therefore, alumina foams can be a suitable material for catalyst preparation. They are used already as filter material and are available at low cost.

According to these promising opportunities, an alumina foam with a porosity of 10 ppi was integrated in the silica glass reactor to investigate the suitability with respect to pressure drop and blockage behavior. The operation of the combustion installation with the foam was carried out in total 25 hours distributed over five experiments. During this time no blockage of the foam could be observed. Though, a higher level of the pressure drop compared to operation without foam was recognizable. An adjustment of the chimney fan was necessary to gain similar results. The pressure difference between operation with and without foam was 5 Pa. Since the use of high temperature stable catalysts is pursued, the foam was integrated at a position, where a temperature of about 970 K was measured during reference operation without foam. The integration of the foam led to a temperature increase of 100 to 150 K at this position. Thus, the foam has a significant influence on the temperature profile in the reactor and acts as heat accumulator. This has to be taken into account for further experiments, when the integration of a catalytic foam at a specified temperature level is necessary to prevent deactivation and ensure optimal catalytic process temperature. The reaction temperature of the catalytic oxidation has to be considered for the right process setup. This is dependent on the catalytic active material, which is selected. Noble metals like palladium and platinum offer higher catalytic activity, but for economic reasons the application of metal oxides or mixed metal oxides including i.e. copper, nickel, iron, cerium, manganese or cobalt are taken into account.

## 4. Conclusion

The investigations show that the developed micro-scale combustion installation offers low energy supply and low pollutant emissions. Downdraught operation as well as continuous fuel supply are key principles of the system and can provide advantages over batch operation systems with upper combustion. Furthermore, basic studies regarding pollutant formation or rather primary emission reduction measures can be carried out. During the first experiments a significant effect of primary and secondary air was observed. CO and VOC concentration below 5 mg/m³ have been measured for two different air supply settings, though for the one with higher primary air rate and lower secondary air rate $NO_X$ concentration was lower. An advancement of the installation is to be carried out to improve the operation of the installation and the reliability of oxygen concentration data. With a view to a future practical use the implementation of gas sensors and a regulation system is considered. Preliminary considerations regarding the integration of a catalyst give a view on a possible emission reduction during maloperation of the installation and the chance for basic research regarding the catalyst integration in biomass combustion systems. The effect of the catalyst in different temperature zones under real operating conditions can be examined and furthermore ageing effects to determine insight regarding the catalyst long-term stability and deactivation mechanism. A promising catalyst concept is the application of alumina based foams.

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