REDUCTION OF NOx FROM COMBUSTION OF BIOMASS, RESEARCH BASED EXPERIENCES FROM EUROPE

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Abstract

Since biomass is the only carbon-based renewable fuel, its application becomes more and more important for climate protection. Among the thermo-chemical conversion technologies (i.e., combustion, gasification, and pyrolysis), combustion is the only proven technology for heat and power production.

Biomass combustion systems are available in the size range from a few kW up to more than 100MW. The efficiency for heat production is considerably high and heat from biomass is economically feasible. Commercial power production is based on steam cycles. The specific cost and efficiency of steam plant is interesting at large scale applications. Hence co-combustion of biomass with coal may be also promising, as it combines high efficiency with reasonable transport distances for the biomass.

However, biomass combustion is related to significant pollutant formation and hence needs to be improved. To develop measures for emission reduction, the specific fuel properties need to be considered. It is shown that pollutant formation occurs due to two reasons:

(1) Incomplete combustion can lead to high emissions of unburned pollutants such as CO and soot. Although improvements to reduce these emissions have been achieved by optimized furnace design including modeling, there is still a relevant potential of further optimization.

(2) Pollutants such as NOx and particles are formed as a result of fuel constituents such as N, K, Cl, Ca, Na, Mg, P, and S. Hence biomass furnaces exhibit relatively high emissions of NOx and sub-micron particles. Air staging and fuel staging have been developed as primary measures for NOx reduction that offer a potential of 50% to 80% reduction. Primary measures for particle reduction are not yet safely known. However, a new approach with extensively reduced primary air is presented that may lead to new furnace designs with reduced particle emissions. Furthermore, assisting efforts for optimized plant operation are needed to guarantee low emissions and high efficiency under real-world conditions.

Key words: *NOx, biomass, fuel, combustion*

I. COMBUSTION OF BIOMASS: FUNDAMENTALS, PRIMARY MEASURES FOR EMISSIONS REDUCTION, AND TECHNOLOGIES

1. Introduction

1.1 Motivation for Energy from Biomass

There is an increasing interest in biomass utilization for energy production worldwide. The driving force for biomass combustion is in most cases either the CO_2 neutrality of sustainable cultivated biomass or the utilization of biomass residues and wastes. Large potentials of both - native biomass as well as biomass wastes - are still available and enable a relevant increase of sustainable bio-energy utilization in the future. Combustion is the most important and mature technology available nowadays for biomass utilization. Improvements with respect to efficiency, emissions, and cost are needed for further exploitation. Beside this, alternatives such as gasification also need to be considered and also combinations of different processes are of interest such as gasification as fuel pre-treatment for co-combustion.

1.2 Feedstock for Biomass Combustion

Combustion can be applied for biomass feed stocks with water contents up to maximum 60%. Fuel constituents beside C, H, and O are undesired since they are related to pollutant and deposit formation, corrosion, and ash. The most relevant constituents in native biomass are nitrogen as a source of NOx and ash components (e.g., K and Cl as a source of KCl) that lead to particulate emissions. Native wood is usually the most favorable bio-fuel for combustion due to its low content of ash and nitrogen. Herbaceous biomass such as straw, miscanthus, switchgrass, etc., have higher contents of N,S, K,Cl, etc., that lead to higher emissions of NOx and particulates, increased ash, corrosion, and deposits. While wood is as well suited for household heating as for larger plants, herbaceous biomass is dedicated for larger plants. The same is true for urban waste wood and demolition wood. The combustion of such contaminated biomass should be strictly limited to combustion plants with efficient flue gas cleaning for the abatement of toxic pollutants such as heavy metals and chlorine compounds.

1.3 Environmental Impact of Biomass Combustion

Biomass furnaces exhibit relatively high emissions of NOx and particulates in comparison to furnaces with natural gas or light fuel oil. Hence, they contribute significantly to particulate matter (PM), ozone, and NO_2 in the ambient air. For wood combustion, a life cycle assessment (LCA) indicates that more than 38 % of the environmental impact of a modern automatic wood furnace is attributed to NOx, 36.5 % to PM 10, only 2 % to CO2, and 22.9% to all other pollutants. The LCA for wood, light fuel oil, and natural gas also shows that the environmental impact of wood is usually higher than that for natural gas for a standard valuation of the greenhouse effect. Hence, improvements in the wood chain are necessary. However, it is also evident that the conclusions of the LCA strongly depend on the valuation of the greenhouse effect since the ranking changes significantly as a result of the different CO_2 impacts of the three fuels.

2. FUNDAMENTALS

Biomass combustion is a complex process that consists of consecutive heterogeneous and homogeneous reactions. The main process steps are drying, devolatilization, gasification, char-combustion, and gas-phase oxidation. The time used for each reaction depends on the fuel size and properties, on temperature, and on combustion conditions. Batch combustion of a small particle shows a distinct separation between a volatile and a char combustion phase with time. For the design of combustion appliances, the high content of volatiles (80 % to 85 %) needs to be respected. For large particles, the phases overlap to a certain extent. Nevertheless, even for log wood furnaces, a certain separation of distinct combustion regimes with time can be found.

Since automatic combustion systems are operated continuously, the consecutive reactions occur simultaneously at different places in the furnace (e.g., in different sections on and above a grate). Hence the zones for different process steps during combustion can be optimized by furnace design. A distinct separation of different process steps can be advantageous with respect to pollutant formation. The main combustion parameter is the excess air ratio that describes the ratio between the locally available and the stoichiometric amount of combustion air.

3. EMISSION REDUCTION

3.1. Staged Combustion

If staged combustion is applied, the excess air can vary in different sections. Two-stage combustion is applied with primary air injection in the fuel bed and consecutive secondary air injection in the combustion chamber. This enables good mixing of combustion air with the combustible gases formed by devolatilization and gasification in the fuel bed. If good mixing is ascertained, an operation at low excess air is possible (i.e., excess air <1.5) thus enabling high efficiency on one hand and high temperature with complete burn-out on the other hand. If good mixing is achieved, the concentrations of unburned pollutants can be reduced to levels close to zero (e.g., CO < 50 mg/m3). However, an accurate process control is needed to ensure optimum excess air in practice. For this purpose, self-adjusting control systems with use of sensors for CO and of CO and temperature have been developed.

3.2 Unburned Pollutants

The main needs for complete burn-out are temperature, time, and turbulence (TTT). The mixing between combustible gases and air can be identified as the factor that is mostly limiting the burn-out quality, while the demands for temperature (around 850°C) and residence time (around 0.5s) can be easily achieved. Sufficient mixing quality can be achieved in fixed bed combustion (two stage combustion). In fluidized bed, good mixing is achieved in the bed and the free board and also dust combustion enables good mixing. For future improvements in furnace design, computational fluid dynamics (CFD) can be applied as a standard tool to calculate flow distributions in furnaces. However, the heterogeneous reactions during drying, transport, devolatilization, and gasification of solid biomass before entering the gas-phase combustion need to be considered as well and needs further improvement to enable the application of whole furnace modeling.

3.3 NOx Emissions

In combustion processes, NO and NO_2 (summarized as NOx) can be formed in three different reactions. Thermal NOx and prompt NOx are formed from nitrogen in the air at high temperatures and in the case of prompt NOx in the presence of hydrocarbons. Further, fuel NOx can be formed from nitrogen-containing fuels. For biomass combustion, fuel-bound nitrogen is the main source of NOx emissions while thermal and prompt NOx are not relevant due to relatively low temperatures. Fuel nitrogen is converted to intermediate components suchas HCN and NHi. These can beoxidized to NOx if oxygen is available, which is the case in conventional combustion. If no oxygen is present, intermediates can interact in the reduction zone and form N_2 in reactions such as $NO + NH_2 = N_2 + H_2O$. During the past 10 years, staged combustion technologies have been developed as a primary measure for process internal NO_x reduction based on this concept, thus leading to the techniques of air staging and fuel staging. Both measures enable a NO_x reduction on the order of up to 50 % for wood with low and up to 80% for bio-fuels with high nitrogen content. However, different specific conditions have to be met accurately to exhaust this reduction potential.

In the case of air staging, a primary air excess around 0.7, a temperature in the reduction zone of 1150°C and a residence time of 0.5s are needed. The relatively high temperature can limit the application in practice due to undesired ash softening and deposit formation. For fuel staging, similar results are achieved at lower temperature, i.e., already at temperatures as low as 850°C. However, the furnace concept and operation is more complex due to the need of two independent fuel feeding systems. For both types of staged combustion, accurate process control is needed to ensure an operation at the excess air ratio needed in the different zones.

Besides primary measures, secondary measures are available for NO_x abatement. The most relevant techniques are selective non catalytic reduction (SNCR) and selective catalytic reduction (SCR) using the same reaction as mentioned for staged combustion, i.e., $NO + NH_2 = N_2 + H_2O$. However, urea or ammonia is injected as reducing agent and as source of NH_2. SNCR has to be applied in a narrow temperature window around 820° C to 940°C, thus enabling a NOx reduction up to 90%. SCR is typically applied in the flue gas in a temperature range around 250° to 450°C and enables a NOx reduction of more than 95%. However, relevant concentrations of undesired side products such as HNCO, N_2O, NH_3, HCN, and others can be formed in both types of secondary measures under unfavorable conditions. Hence, primary measures are preferable if they can achieve sufficient emission reduction.

3.4 Particulate Emissions

Biomass combustion leads to relatively high emissions of particulates, i.e., well above 50mg/m3 at 11 vol % O_2. The majority of the particulates are smaller than 10 µm with a high share of sub-micron particles. The composition of sub-micron and super micron particles in fluidized bed combustion is distinctive as the fine particles are composed mainly of K, Cl, S, Na, and Ca and the coarse particles of Ca, Si, K, S, Na, Al, P, and Fe. In fixed bed combustion, increasing mass concentrations of particulate emissions are typically related to increasing mean diameter. Further, a dependency of the particle composition on size can also be found in fixed bed conditions, as K, S, Cl, and Zn are mainly found in the sub-micron fraction, while the content of Ca is increasing with increasing particle size. If almost complete burn-out is achieved by appropriate furnace design, the particulates result almost exclusively from ash components in the fuel with salts such as KCl as main components. The main fuel constituents with respect to aerosol formation are typically K, Cl, S, Ca, Na, Si, P, Fe, and Al. Primary measures which can safely meet a high reduction potential, i.e., by at least a factor of 10, of this category of aerosols are not known so far.

A new approach for primary particle reduction has been presented recently. It was shown, that particles from wood combustion are mainly formed by nucleation, coagulation, and condensation during temperature decrease in the boiler. Further, these particles are mainly salts and consist mainly of K. K in the fuel is present as a salt with high melting point and devolatilization temperature. If oxygen is available at high temperature, a high share of K can be oxidized. As K oxides have significantly lower devolatilization temperatures than the K salts, they are almost completely vaporized into the gas phase and lead then to particle formation from the gas phase. If no oxygen is present in the fuel bed, the conversion of K to volatiles may be reduced since the majority of K salts can be converted into the grate ash. Since a similar behavior for other ash components in the fuel is assumed, the oxygen content during the solid fuel conversion is regarded as a key parameter for aerosol formation. As the results from the Swiss laboratory furnace show, a reduction of particle emissions in the order of a factor of 5 is achievable, i.e., from160-195 mg/m3 to 20 - 45mg/m3 (13%O_2). With most of the investigated fuels, particle emissions below 50mg/m3 (13%O_2) were achieved. Since fuel composition is the main parameter for aerosol formation, secondary measures are necessary as well for further reduction in the future. Among those, fabric filters are regarded as most promising. However further developments and adaptations are necessary for particle removal technologies appropriate for the needs and size for typical biomass combustion at reasonable cost.

4. CONCLUSIONS

Biomass combustion contributes significantly to the global energy supply nowadays. It is a proven technology and widely applied in the size range from a few kW for household heating to several MW for district heating and up to more than one hundred MW for power stations based on steam cycles. However, there are strong needs to improve biomass combustion in terms of environmental impact. Although the measures for complete combustion are well-known in principle, future efforts for improvement are needed to implement sufficient gas mixing in commercial furnaces and to guarantee correct operation in practice. Combustion modeling including CFD can be applied for further optimization of furnace design in future. Further, conversion efficiencies from biomass combustion to heat still have a potential for improvement. On one hand, applications in practice have a high potential of improvement by system optimization, improved maintenance correct adjustment of fuel-to-air ratio, and optimized management of heat distribution. On the other hand quality assurance for the new realization of complex heating plants is recommended.

The key parameter for high combustion efficiency is the excess air ratio, which in most applications is far higher than theoretically and practically needed. To enable operation at optimum excess air and guarantee maximum efficiency in practice, advanced process control should be adapted for the large variety of applications and widely implemented in the future. For the combustion of biomass with high water content, process economy can be improved either by flue gas condensation (without heat-consuming vapor abatement) and/or process integrated fuel drying with waste heat.

The remaining issues after implementation of these techniques are the reduction of NO_x emissions and the reduction of particles that are mainly in the sub-micron size range. The most promising technology for further emission reduction is staged combustion including air-staging, fuel staging and advanced staged combustion at extremely low primary air. However, since both pollutants result predominantly from fuel constituents, i.e., fuel nitrogen and ash components respectively, there are physical limitations to reducing NOx and particles by primary measures, since high temperatures as well as the presence of oxygen are system immanent in combustion. Following, there are two options, which have to be considered for the future as well. On one hand, secondary measures need to be developed for the specific requirements of biomass combustion. Although certain technologies for NOx and particulate abatement exist, they are not yet properly implemented (e.g., air staging and fuel staging) or they do not fulfill the needs for biomass for reasonable cost in the most frequently used appliances up to 5MW.

II. FUEL STAGING FOR NOx, REDUCTION IN BIOMASS COMBUSTION: EXPERIMENTS AND MODELLING

With increasing use of biomass in combustion processes, the reduction of the related NO_x emissions which originate mainly from the fuel nitrogen becomes more and more important.

Efficient primary measures for NO_x reduction are staged combustion techniques. Air staging has been investigated earlier and has found its way into practice. Since fuel staging has been only very rarely applied with non-pulverized biomass yet, the aim of the present work was to investigate the potential of fuel staging for NO_x reduction in fixed bed systems.

For this purpose, a prototype under-stoker furnace of 75 kW thermal input with two fuel beds in series was developed. Experiments were performed with wood chips with low nitrogen content, and a chipboards with high nitrogen content to investigate the influences of the main process parameters, i.e., stoichiometric ratio, temperatures, residence time, and fuel properties on the conversion of fuel nitrogen to N-species.

The most important parameters were found to be the temperature and the stoichiometric ratio in the re-burn zone. The potential of fuel staging was measured and compared with air staging and un-staged combustion. The experiments show that low NO_x emissions are already achievable with fuel staging at lower temperatures than with air staging, i.e., 900-1000 C, and at a stoichiometric ratio of 0.85 in the reduction zone.

The combustion of natural wood chips in current firing system lead to emissions of NOx < 200 mg/m³ and Particulate Matter (PM) < 50 mg/m³. During the combustion of chipboard residues NO_x emissions reached 1000 mg/m³ and PM 500 mg/m³. The NO_x reduction achieved under optimum conditions for the chipboard as main fuel was 78% which is higher than with air staging, where 72% NO_x reduction was measured. For wood chips both measures attained about 66%.

The nitrogen conversion during air and fuel staging has also been simulated using a furnace model based on ideal flow patterns as perfectly stirred reactors and plug flow reactors. A detailed reaction mechanism including the nitrogen chemistry was implemented. The trends found with this model are in good agreement with the experiments and they indicate that even higher NO_x reduction may be reached with improved process design. The investigations show that fuel staging is a promising technology for NO_x reduction also for fixed bed biomass furnaces.

REFERENCES

Thomas Nussbaumer, Verenum, Langmauerstrasse 109, 8006 Zurich, Switzerland
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