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## SYSTEMS FOR FLUE GASES TREATMENT AT THE COMBUSTION OF (AGRICULTURAL) BIOMASS

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**Abstract.** *Biomass combustion has many advantages compared to fossil fuels due to reduction of carbon dioxide emissions, but several issues are present during biomass combustion. One that must always be considered is emission of nitrogen oxides (NO<sub>x</sub>), which have significant impact on the environment and health. At biomass combustion, main mechanism responsible for NO<sub>x</sub> emission is oxidation of fuel-bound nitrogen. Increased NO<sub>x</sub> emissions are particularly present at agricultural biomass combustion, due to its increased nitrogen content, which is further expressed by the use of nitrogen fertilizer. Denitrification methods are classified as pre-combustion, combustion control and post-combustion (flue gas treatment) methods. Aim of this paper is to provide insight in different flue gas denitrification methods (DeNO<sub>x</sub> measures) from the review of existing literature, with emphasis on post-combustion methods, since they have higher efficiency. Also, pre-combustion and combustion control methods at agricultural biomass combustion often do not provide reduction below emission standards. Selective catalytic (SCR) and selective non-catalytic reduction (SNCR), wet scrubbing, adsorption, electron beam, electrochemical method, non-thermal plasma and microbial approach were considered. For each method, mechanism of NO<sub>x</sub> reduction is given and discussed, alongside with efficiency that could be achieved. Overall comparison of advantages and disadvantages for these methods is provided. Further research of denitrification and optimization of described methods, to overcome NO<sub>x</sub> emission problem is required. Application of these methods in commercial use and increasing their efficiency, while solving cost and methods disadvantages, is key for biomass to be used as a renewable energy source and fossil fuel replacement.*

**Key words:** *biomass, combustion, nitrogen oxides (NO<sub>x</sub>), denitrification, selective catalytic reduction (SCR), selective non-catalytic reduction (SNCR)*

### 1. INTRODUCTION

Biomass combustion has many advantages compared to fossil fuels due to the reduction of carbon dioxide emissions and can be considered a renewable energy source

if used properly. Since biomass is part of the Earth's carbon cycle it can be assumed that biomass is almost carbon neutral. One of the issues present during biomass combustion is the emission of nitrogen oxides (NO<sub>x</sub>). It is not possible to reduce carbon monoxide, carbon dioxide, hydrocarbon emissions and nitrogen oxides at the same time. This is particularly pronounced with the use of fuel containing a high percentage of bound nitrogen, such as biomass feedstocks (plants, wood, waste). When examining the impact of CO<sub>2</sub> on NO formation it was concluded that the NO release was increased with char conversion for a gas mixture of CO<sub>2</sub>/O<sub>2</sub>, while for H<sub>2</sub>O/O<sub>2</sub> the release decreased 1.. In general from agriculture biomass combustion the pollutant emissions can be divided into unburnt pollutants (CO, PAH, HC, C<sub>x</sub>H<sub>y</sub>, PM, tar and char particles) and pollutants which emissions are influenced by fuel characteristics. Alongside NO<sub>x</sub> (NO and NO<sub>2</sub>) in the second group of pollutants is an emission of nitrous oxide N<sub>2</sub>O as well 2.. Compared to NO<sub>x</sub>, the emission of N<sub>2</sub>O is significantly lower and is usually not taken into consideration in standards and emission limits. N<sub>2</sub>O is not only direct greenhouse gas but also has an indirect effect on the depletion of the ozone layer 3.. Recently more attention is given to this matter 4.. In this paper, only NO<sub>x</sub> emission is taken into consideration. NO<sub>x</sub> harm the human respiratory system, they affect O<sub>3</sub> formation, vegetation damage, photochemical smog formation, etc. 5,6.. Increased NO<sub>x</sub> emissions are particularly present at agricultural biomass combustion, due to its increased nitrogen content, which is further expressed by the use of nitrogen fertilizer. The steam, gas and electricity supply are the largest source of NO<sub>x</sub> emission in Serbia (fig. 1) 7..

Formation mechanisms for NO<sub>x</sub> 8.: 1) Thermal mechanism – oxidation of the molecular nitrogen with oxygen in combustion air, present at high temperatures (above 1300°C); 2) Prompt mechanism – fast reactions in the flame zone of the fuel-bound hydrocarbons radicals and nitrogen from the atmosphere (N<sub>2</sub>) at low temperatures and fuel-rich conditions; 3) Oxidation of fuel-bound nitrogen (fuel-N). At biomass combustion, the main mechanism responsible for NO<sub>x</sub> emission is the oxidation of fuel-N. Temperature, residence time, mixing conditions, and excess air ratio affect NO<sub>x</sub> formation and reduction as well. Thermal-NO and prompt-NO can be neglected for agricultural biomass (low-temperature combustion, below 1000 °C). The catalytic effect for NO<sub>x</sub> reduction can be provided by char and ash. Since agricultural biomass generally contains high contents of volatile matter and low contents of fixed carbon, the effect of char on the NO<sub>x</sub> and N<sub>2</sub>O formation is usually insignificant. The catalytic effect of the ash is important for residues with high CaO/alkaline oxides contents (mustard husk, cotton husks, groundnut husks, etc.). Simultaneous reduction of CO and NO<sub>x</sub> is very problematic 8-11.. This paper aims to provide the importance of NO<sub>x</sub> emissions reduction and give a review of existing and promising methods in development.

## 2.DENITRIFICATION METHODS

According to various authors 10,12., denitrification methods can be classified as pre-combustion, combustion-control and post-combustion methods. Pre-combustion and combustion control methods at agricultural biomass combustion often do not provide reduction below emission standards, so the emphasis is on post-combustion methods since they have higher NO reduction efficiency for this type of fuel.

**Pre-combustion methods** include: proper selection of fuel (biomass), pretreatments that affect reducing heterocyclic-N compounds 10., application of fuel additives, biomass fuel blending and co-combustion with fossil fuels.

**Combustion-control (primary measures)** refer to a modification of the combustion process or operating conditions on existing furnaces without the use of additional pollution abatement equipment behind the main combustion zone. Since the biomass is being burned, strategies for modification of operating conditions that can be applied are reduction of oxygen concentrations in the combustion zone and improving mixing conditions. Modifications of the biomass combustion process that are applicable for NO<sub>x</sub> reduction are fuel staging, flue gas recirculation and low NO<sub>x</sub> burner. **Air staged combustion** provides good mixing conditions (combustion air and combustible gases from gasification and devolatilization) 17.. During two-stage combustion primary air is injected into the fuel bed, secondary air is successively injected in the combustion chamber. This way fuel-rich primary and fuel-lean secondary zones are created. It is required that the primary air is under stoichiometric condition (0.7-0.9) since it causes nitrogen from volatiles to form nitrogen containing NO<sub>x</sub> precursors (NH<sub>3</sub>, HCN, HOCN, NO), than in the second stage sufficient air is added, to achieve complete burnout, and it enables NO reduction. NO<sub>x</sub> here acts as an oxidant agent for nitrogen containing emissions and products of incomplete combustions (NH<sub>2</sub>, NH<sub>3</sub>, CO, etc.) 9.. With the application of **fuel-staging** fuel is supplied to the furnace at two levels. Primary fuel is combusted with excess air above 1. Secondary fuel is fed with the last portion of the air above the combustion zone, creating a fuel-rich secondary combustion zone in which NO<sub>x</sub> from the primary fuel-lean zone is reduced 9.. **Flue gas recirculation (FGR)** affects flame temperature reduction and excess air. Recirculated flue gas is considered an inert gas mainly of CO<sub>2</sub>, H<sub>2</sub>O with low concentrations of products of incomplete combustion. FGR act as NO<sub>x</sub> reducing agents. **Low NO<sub>x</sub> burners** are used in utility boilers for steam and electricity production, so they are considered for coal and biomass co-combustion 18..

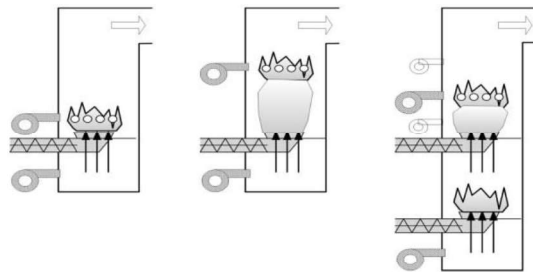


Fig. 2 Combustion: conventional, air staged and fuel staged (left to right) 3.

**Post-combustion methods (secondary measures) – Flue gas treatment** include two main approaches: 1) NO<sub>x</sub> removal from flue gas (absorption or adsorption processes); 2) NO<sub>x</sub> destruction (transformation of NO<sub>x</sub> into non-toxic products) 19.. For the first approach additional medium is needed, also in most cases waste is generated and appropriate treatment is required.

### 3. FLUE GAS TREATMENT

**Selective catalytic reduction (SCR)** is a technique of NO<sub>x</sub> conversion into N<sub>2</sub> and H<sub>2</sub>O vapor with catalyst support. The reductant is being added to a stream of flue gases by injection before reaching catalyst placement. In reactions of a mixture of flue gases and reducing agent with catalyst, NO<sub>x</sub> are being removed. System arrangement is shown in the scheme in fig. 3. Reducing agents that are currently in use are ammonia (NH<sub>3</sub>-SCR) or urea, hydrocarbons (HC-SCR), hydrogen (H<sub>2</sub>-SCR), carbon monoxide (CO-SCR). Reducing agent NH<sub>3</sub>-SCR can be used in liquid form as anhydrous ammonia or aqueous ammonia. Hydrogen (H<sub>2</sub>) is considered a clean, non-toxic reducing agent compared to ammonia (no secondary pollutants). H<sub>2</sub>-SCR proceeds at relatively low temperatures. Difficulties that come along with H<sub>2</sub> usage are transportation and the formation of explosive mixtures in presence of oxygen 20.. Hydrocarbons (HC) are characterized by practicality and cost-effectiveness. Unburnt HC from exhaust gas can be used (especially promising in the automotive industry). The problem with HC is their composition of various alkanes, alkenes and alkynes, which leads to a more difficult prediction of their behavior through the NO<sub>x</sub> reduction. This occurs because the effect of the mixture is different than when they are being used individually 21.. H<sub>2</sub>-SCR and HC-SCR show lack of reacting selectively with NO<sub>x</sub> in flue gases (more likely they will react with O<sub>2</sub>). Besides, at higher temperatures (>500°C) HC are consumed in combustion reactions. As a result, these reducing agents are applicable at lower temperatures (<200°C) on supported noble metals (Pt and Pd), which drastically increases the cost of their application. Contrary to that, ammonia and urea can be used at higher temperatures, and regardless of the presence of oxygen, they prefer to react with NO<sub>x</sub>. Urea is being used as crystal granules dissolved in water 10. Carbon monoxide (CO), as one of the contaminants in the combustion process, is more available and has a more affordable price than NH<sub>3</sub>. As a reducing agent, CO gives good results in NO<sub>x</sub> reduction and gives the possibility of simultaneous extraction of NO and CO from flue gas 22.. This method also requires the development of specific low-temperature catalysts and is still in the testing phase. Catalyst is necessary for SCR to be functional and effective, it should possess high selectivity and activity for N<sub>2</sub> and poisoning resistance. It can be made of a single component, a combination of multiple components, or an active phase. Reduction occurs in catalyst activated sites, which are practically catalyst's pores. For different operating conditions various catalysts have been developed: form noble metals, supported noble/transition metals, supported transition metals oxides, transition metals, metal oxide, zeolites, mixed oxide catalysts, carbon materials, etc. Noble metals are non-toxic and clean as reducing agents and when used formation of secondary pollutants doesn't occur. They have poor resistance to sulfur dioxide and are highly costly. Copper-based catalysts have low cost, they are characterized by, stability, catalytic activity, and selectivity 6., 20.. In addition to the above, catalysts differ in catalytic activity temperature domain, and therefore have different locations in combustion systems, which can be seen in more detail in 10.. Problems that could arise are the deactivation and contamination of catalysts from fly ash particles and aerosols, as well as N<sub>2</sub>O formation as a by-product of reactions on SCR catalyst 6..

**Selective non-catalytic reduction (SNCR)** is based on a chemical reduction of NO<sub>x</sub>, which occurs when the is injected into a stream of hot flue gas. In the reaction of the reagent with flue gas, NO<sub>x</sub> are being converted into N<sub>2</sub> and H<sub>2</sub>O vapor. System

arrangement is shown in the scheme in fig. 4. NO<sub>x</sub> reduction is achieved due to high temperatures, at which reagent tends to react with NO<sub>x</sub> rather than with other components from flue gas, hence the name selective non-catalytic reduction.

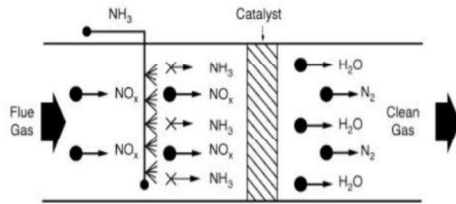


Fig. 3 SCR 10.

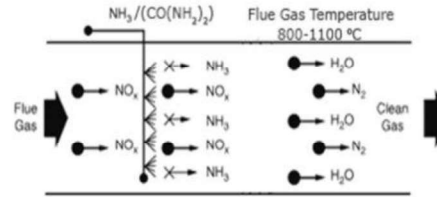


Fig. 4 SNCR 10.

Reagents that are currently in use for SNCR are ammonia (NH<sub>3</sub>-SNCR), urea (CO(NH<sub>2</sub>)<sub>2</sub>) and cyanuric acid (C<sub>3</sub>H<sub>3</sub>N<sub>3</sub>O<sub>3</sub>). Among reducing agents urea is most environmentally friendly due to non-toxicity, therefore storage is easier. The main drawback for urea usage is higher cost and possible increased N<sub>2</sub>O generation during reduction 18.. From the literature review, the following can be concluded for SNCR. It is crucial to accomplish the temperature window for all operating conditions and boiler loads (at temperatures >1100°C decomposition of reactants occurs, and at < 800°C there is no reaction of NO reduction at all). There are variations in the temperature of flue gases along with the height of the furnace, which is a consequence of the variable composition and calorific value of biomass and deposits formation on the exchange surfaces (in the latter case, as a rule, the temperature window shifts upward). The temperature range depends on the reagent being used, as well as the ratio of NO<sub>x</sub> from flue gas and reagent. Since reagents have different optimal temperature ranges, reagent replacement for the same system to adapt to the current temperature window is a possible solution. Choosing an appropriate place for reagent injection is important, to avoid uneven distribution across the boiler cross-section. Multilevel injection location, as well as prediction of the additional ones, are important. If this is not satisfied, the so-called ammonia slip happened, which results in higher NO<sub>x</sub> and excess NH<sub>3</sub> at the end of the process. The enhancers (also called additives) can be used for lowering the temperature range for SNCR: CO, H<sub>2</sub>, CH<sub>4</sub>, a combination of hydrocarbons, ethylene glycol, furfural, hexamethyl tetra-amine, syngas, sodium compound, and CH<sub>3</sub>NH<sub>2</sub> 20.. Their influence on NO<sub>x</sub> reduction is realized due to radicals for the initiation of the reaction, that they provide. Residence time has a significant impact on NO<sub>x</sub> reduction with SNCR and depends on the temperature range that is applied. When stricter NO<sub>x</sub> reduction is required hybrid SNCR-SCR system can be applied (SCR in the duct). When ammonia as a reagent is used, from SNCR ammonia slip is purposely generated to feed SCR catalyst. Reduction of NO<sub>x</sub> to 90% with reduced SCR catalyst volume is possible with hybrid system application.

**The wet scrubbing** technique has the ability of simultaneous NO<sub>x</sub> and SO<sub>x</sub> reduction and is considered an environmentally benign method. As by-products N<sub>2</sub>O, N-S complexes and NH<sub>3</sub> are formed during the reduction of NO<sub>x</sub> and SO<sub>x</sub>. In the reduction process, NO<sub>2</sub> and other water-soluble gases as CO<sub>2</sub> and SO<sub>2</sub> pass through alkali solutions where their transformation in acids occurs. Afterward, the process of neutralization is

used. Removal of NO is more complicated because of lack of water- solubility, so additives are necessary 23.. As a solvent freshwater is mostly used. 24. have researched electrolyzed seawater usage for NOx and SOx removal in semi-continuous bubble column reactor using scrubbing system. With an increase of active Cl concentration of electrolyzed seawater NOx reduction efficiency increased as well. Initial pH has a significant impact on NOx removal, if it is in a convenient range process is less sensitive to inlet NO concentration and reaction temperature. Different aqueous solutions are used: sodium chlorite (NaClO<sub>2</sub>), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), potassium permanganate (KMnO<sub>4</sub>), urea solution (CO(NH<sub>2</sub>)<sub>2</sub>), ozone (O<sub>3</sub>), (NH<sub>3</sub>/NaClO), etc. In combination with Fe, Co, Cu and Ru are used different chelated compounds as ethylenediaminetetraacetate (EDTA), nitrilotriacetic acid (NTA), dimercaptopropanesulfonic acid (DMPS), citrate and diethylenetriamine pentaacetic acid (DTPA). Other absorption systems include activated carbon fibers (ACF), zero-valent iron (ZVI), FeO, fly ash, microalgae and fungi.

**Electron beam** flue gas treatment is also a method for simultaneously NOx and SOx removal. Before irradiation ammonia is being injected into the flue gas stream. By irradiation of flue gas with fast electrons, chemical changes are initiated. They make NOx and SOx removal easier. Then in the interaction of fast electrons and flue gas components (N<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>O, CO<sub>2</sub>) oxidants, like those produced by UV sunlight are being produced, with a difference at concentration levels several orders of magnitude higher. In the reaction of these oxidants with NOx and SO<sub>2</sub> nitric and sulfuric acids are formed, which create a solid powder of ammonium nitrate and sulfate. Lastly, by-products are being filtrated. A generated by-product is a mixture of ammonium sulphate and nitrate. 25.

With the **electrochemical method (EM)**, NOx from flue gas are being converted into harmless gases (N<sub>2</sub> and O<sub>2</sub>), without waste as spent scrubbing solution. NOx reduction occurs on the cathode when potential is applied to the solid-state electrochemical cell. At the cathode N<sub>2</sub> and O<sub>2</sub><sup>-</sup> (oxide anions) are formed from NOx. Through the electrolyte oxide anions are being transported to an anode where oxygen formation occurs. For NOx reduction in an oxygen-rich environment, the cathode of an electrochemical cell must have high selectivity towards NOx so that oxidation reaction does not prevail. Adding NOx adsorption materials (alkali metal oxides or alkaline earth metal oxides) to the cathode affects the increase of its selectivity. 26.

**Non-thermal plasma (NTP)** can be induced by: electron beam irradiation, pulsed corona discharge, dielectric barrier discharge and as microwave plasma. Reduction of NOx and SO<sub>2</sub> with NTP is almost simultaneous, takes place at atmospheric pressure, and it occurs due to reactions with free radicals (H, N, O, OH, O<sub>3</sub>, HO<sub>2</sub>, etc.). Firstly the energetic electrons in plasma collide with H<sub>2</sub>O, N<sub>2</sub> and O<sub>2</sub> to form gas-inducing primary radicals (HO, O, N) and ions. Excited molecules, like oxygen, in rapid reactions with the main gas quench and generate more HO and O radicals. More secondary radicals are being produced in reactions of the electron-ion and ion-ion and possessing higher energy than gas molecules. In the oxidation of NOx and SO<sub>2</sub> by these radicals, HNO<sub>3</sub> and H<sub>2</sub>SO<sub>4</sub> are being formed almost instantly (<10<sup>-3</sup> s). The obtained products are neutralized using NH<sub>3</sub>. As a result, NH<sub>4</sub>NO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> are produced. It is a new and promising deNOx technique that has importance especially in the automotive industry as a hybrid method with HC-SCR. 27.

**Microbial approach** is a novel alternative treatment method involving the usage of autotrophic microorganisms, which are capable of making their own food in the transformation of inorganic substances into organic nourishment. Inorganic sulphides are being used as electron donors, NO<sub>x</sub> reduction occurs through biochemical reactions and at the same time microorganisms are being fed and can grow. The whole process can be divided into the dissolution phase, in which SO<sub>2</sub> and NO<sub>x</sub> are being transferred to the liquid phase while passing the gas-liquid double membrane, and the biochemical phase in which they are consumed by microorganisms. Most of NO is being adsorbed on the surface of microbial cells. A small amount can dissolve in the solution and can be oxidized into nitrates (by denitrifying microorganisms) or reduced to N<sub>2</sub> (nitrogen reductase). NO<sub>2</sub> is reduced to N<sub>2</sub> after being converted to nitrate/nitrite (nitrogen reductase). 12. This method merits further study because according to the available literature, all tests were performed at temperatures below 60°C, so that at this stage of development they can only be used for further treatment of denitrification by-products, such as at wet scrubbers 28..

#### 4. CONCLUSION

Although the problem of emission of nitrogen oxides is not new, it is still being investigated for an efficient solution to be found. In addition to improving conventional methods, new methods and approaches are being developed. There is no general solution for this problem, it is necessary to adapt to the existing conditions and to choose the reduction method that is most optimal for the considered system. This especially refers to modified systems that have switched to working with biomass. In the table 1 is given overview for reviewed methods alongside with their main advantages and disadvantages. Further research of deNO<sub>x</sub> and optimization of described methods, to overcome NO<sub>x</sub> emission problem is required. Application of these methods in commercial use and increasing their efficiency, while solving cost and methods disadvantages, is key for biomass to be used as a renewable energy source and fossil fuel replacement.

Table 1: Overview for reviewed methods

Method	SCR	SNCR	Wet scrubbing	Electron beam	EM	NTP	Microb. approach
Efficiency %.	60-90	30-70		70-90	approx. 60		40-90
Temp. range °C.	150-590	800-1100	Ambient temp.		400		> 60
Advantages	High efficiency Good selectivity Cost effectiveness Simple system Mature method	Simple solution Implementation on existing systems is easy Lower cost Combination with other deNOx methods Mature method	PM and acid gases removal No high temp. No extraneous chemicals Easy regeneration Low cost of equipment and additives Mature method	No waste generation By-product can be used for fertilizer production	Reductant is not required Simple, clean and compact technology	Simple system and operation No waste generated By-product are useful and can be used for fertilizer production	Reduction of CO <sub>2</sub> Less secondary pollution Low energy consumption High removal rate No need for catalysts or chemicals
Disadvantages	High cost, lifespan and disposal of catalyst Storage of reducing agents High cost and corrosion of equipment Ammonia slip	Less reduction Downstream equipment cleaning is necessary High operating temp. Ammonia slip	Slow removal rate Low efficiency Liquid waste	High energy consumption Prevention of radiation problems Complex equipment Not tested in real-scale systems	Still not developed and implemented	Low efficiency High cost and energy consumption Reactor complexity Cost and lifetime of the plasma power supply Physicochemical property change of by-products	Unstable removal efficiency Short lifespan of microorganisms Insufficient temp. for FGT High sensitivity to temp. fluctuations



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