

SNCR in biomass combustion facilities: from theories to existing models

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With the significant use of biomass for energy purposes, as the only CO₂-neutral carbon-based renewable energy source, besides several well-known advantages, certain disadvantages appear. One of them is the possible increased emission of nitrogen oxides, NO_x, which is especially pronounced with the usage of agricultural biomass. Given the available resources of this biomass and increasingly stringent legal norms regarding NO_x emissions, this problem needs special attention, which is the topic of this paper. The paper presents: mechanisms of NO_x formation and conversion of nitrogen from fuel during combustion, a brief overview of available methods for denitrification, both conventional and newer ones that have not yet come to industrial-scale application. NO_x emissions from biomass combustion are mainly caused by fuel-bound nitrogen, which is especially present in biomass with a high content of bound nitrogen such as agricultural biomass. Therefore, the focus is on selective non-catalytic reduction, SNCR, as a secondary measure of denitrification. Theoretical bases of SNCR, presentation of SNCR reagents, main pathways of chemical reduction reactions for different reagents, and reaction kinetics are given. Also, the general bases for setting numerical SNCR reduction models and reviewing existing ones, as well as experimental results from the available literature, are presented. Finally, examples of industrial applications of SNCR reactors and general recommendations for the application of this denitrification measure are introduced.

Keywords: SNCR, biomass combustion, NO_x

INTRODUCTION

After the industrial revolution and the damages caused by the use of fossil fuels, the humanity is facing various challenges associated with sustainable development, the transition to green energy sources, as well as reducing greenhouse gas emissions and planning the formation of self-sustaining cities. When choosing an energy source, one of the main conditions is that it is possible to respond to consumer demands throughout the year. From the point of view of sustainable development, the residues from agriculture are especially significant. In order to work on the introduction of biomass in the energy sector, it is necessary to observe the shortcomings that come with it in time. An inseparable problem related to the biomass combustion is the emission of nitrogen oxides NO_x (nitrogen monoxide (NO) almost 90%, and nitrogen dioxide (NO₂) <10%), which have extremely negative effects on human health and the environment in general [1, 2]. Due to combustion conditions, the predominant influence on the mechanisms of NO_x formation, which are thermal, prompt and fuel-N, has the last one, which refers to the oxidation of nitrogen bound in the fuel [3]. Since fuel-N mechanism mostly depends on the fuel composition, and not optimal combustion conditions

it is not possible to reduce NO_x emissions without secondary denitrification measures [4]. Various technologies have already been developed and many are still being researched. Neither of them can be singled out as the best so the selection should be made according to techno-economic analysis which should provide the highest efficiency with the lowest cost. Selective non-catalytic reduction (SNCR) is considered as a possible solution to all of the previously mentioned problems. One of the advantages of SNCR is that it can be easily combined with other denitrification measures. For a technology to be well understood, a comprehensive approach is needed, from the initial theory to the implementation in reality. This paper presents a concise guide through SNCR denitrification method, and refers to the NO_x formation during biomass combustion, comparison of SNCR with the other methods, influential parameters based on theoretical and experimental studies, detailed chemical mechanisms, numerical modeling, as well as the challenges associated with them.

METHODS

SNCR-associated studies that meet the inclusion criteria are observed. The first requirement concerns the type of fuel – biomass with high nitrogen content. This refers to flue gas from the combustion

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of biomass or synthetic gas, which should correspond to the composition of flue gases in biomass. Alongside that, thermal and prompt mechanisms of NO_x formation are neglected. They are not significantly expressed in biomass because of the relatively low combustion temperature due to the low melting temperature of biomass ash [5]. By observing the temperature range of biomass combustion, it was decided to consider ammonia and urea as reducing agents. A selection of concepts and ideas that are important for a comprehensive understanding and analysis of SNCR has been made. Based on them, previous research has been classified into sections that can be considered necessary steps for the introduction of SNCR measure into the energy and industrial sector. To make the review comprehensive, not only the latest studies were included.

RESULTS AND DISCUSSION

Firstly, it is necessary to understand the conversion of nitrogen during biomass combustion and NO_x formation, which is explained in the first section. After that, it is significant to consider comparing SNCR technology with other denitrification measures that are available and under development. Afterward, it is possible to dedicate to the fundamental theoretical knowledge and the most influential parameters of the SNCR. That also includes conclusions obtained from experimental research. The following sections refer to detailed chemical mechanisms and numerical modeling of the reduction process. For each section, the fundamental principles established so far, current issues, as well as the next steps needed for further development of the SNCR, are observed. Finally, the emission of nitrogen oxides from real-scale biomass combustion systems is presented, with and without SNCR measure, which shows the reduction in reality. That together provides the necessary prior knowledge for monitoring the application of SNCR in plants, predicting NO_x emissions, along with compliance with existing legislation.

Conversion of nitrogen from fuel (fuel-N) during biomass combustion (N paths)

To solve the NO_x emission problem, first, it is necessary to understand the process of NO_x formation during biomass combustion. Depending on the fraction of volatiles and char (whose fraction in the fuel can go up to 30% [6, 7]), significant deviations in the process can occur. Besides the fraction of volatiles and char, important parameters are also temperature, time of the combustion process, and size of the particles that are burned. In

addition to the previous parameters, the influence and presence of H₂O, ash, and various compounds that appear in the fuel composition are considered. Nitrogen conversion during combustion is differentiated by combustion phases, although in reality, these phases overlap [8, 9]. Fig. 1. shows a schematic diagram of the nitrogen conversion pathway in biomass combustion, from the main forms in the fuel (mostly proteins) [10, 11], to the combustion chemical species and nitrogen remaining in the ash.

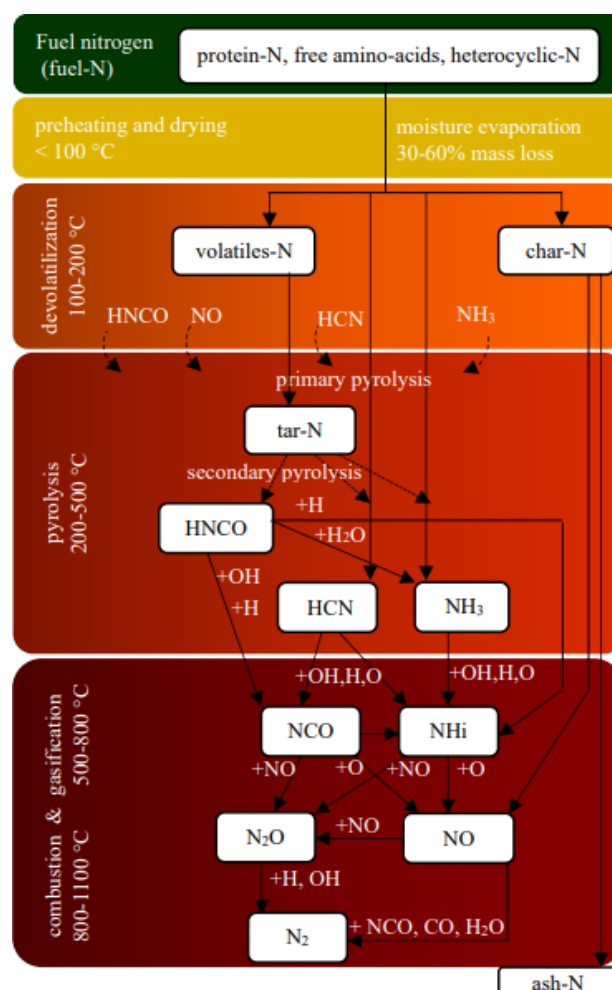


Fig. 1. Conversion of fuel-N (biomass combustion)

NO_x formation is not uniform during biomass particle combustion. Two peaks occur, one due to the volatile-N [4] and the other due to the char. Amongst the chemical species that can be found during biomass combustion, it is important to single out NH₃ and HCN, which have a crucial role in the formation of NO_x [12, 13]. The majority of them are formed during the tar cracking in the secondary pyrolysis [10]. The ratio of these precursors depends on the operating conditions of the process and the type of combustion - a difference can be noticed when observing the combustion of individual biomass particles compared to the combustion in the

real system. It was found that when the initial nitrogen content increases, both the HCN / NH₃ ratio and the temperature decrease [14]. The content of H₂CO should also be considered; even though it is present in a smaller amount, it still contributes to the formation of NO_x [15, 16].

The process of NO_x reduction is highly affected by the char structure [17] and the presence of ash which can have a catalytic effect on NO_x reduction. This is especially important in agricultural residues with a high content of CaO / alkali oxides [5]. Out of all nitrogen conversion pathways, volatiles have a dominant influence, and the possibility of predicting NO_x formation from them is particularly significant [18–20].

Comparison of deNO_x measures

This section aims to give an objective comparison of the available measures that correspond to the temperature range of biomass combustion. According to various authors, denitrification methods can be classified as pre-combustion, combustion-control, and post-combustion methods (flue gas treatment). Combustion-control (primary measures) refers 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. Modifications of the biomass combustion process refer to air and fuel staging, flue gas recirculation, and low-NO_x burner. Strategies for modification of operating conditions that can be applied during biomass combustion are reduction of oxygen concentrations in the combustion zone and improving mixing conditions [21–23]. Pre-combustion and combustion control methods at agricultural biomass combustion often do not provide reduction below emission standards so post-combustion methods are necessary. In Table 1 a comparison of measures is given. Further research of deNO_x and optimization of described methods, to overcome the NO_x 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.

SNCR in brief

Selective non-catalytic reduction (SNCR) is based on a chemical reduction of NO_x, which occurs when it is injected into a stream of hot flue gas. In the reaction of the agent with flue gas, NO_x are being converted into N₂ and H₂O vapor (and CO if urea is used). NO_x reduction is achieved due to high

temperatures, at which the agent tends to react with NO_x rather than with other components from flue gas, hence the name selective non-catalytic reduction. The absence of catalysts and all problems related to it is a great advantage of this deNO_x method. In principle, the reduction process takes place due to chain reactions maintained by free radicals, which are provided by reduction agents. It has already been mentioned that during nitrogen conversion during combustion, the formed NO_x can be partially reduced to N₂, which can be seen in Fig. 1. The goal is to increase the concentration of free radical-producing compounds that are essential for NO_x reduction, within a temperature range that provides selectivity so that the reduction is not replaced by further NO_x formation. Ammonium and urea as reducing agents are effective at high temperatures due to their decomposition to NH₂ which is further explained in the section *Detailed chemical mechanism*.

1. Comparison of reducing agents.

Ammonia and urea are known as reducing agents for biomass. Some studies consider the possibility of other agents, i.e. methane [24], hydrazine hydrate, urea-spiked hydrazine [25], and cyanuric acid–RapDeNO_x, but they are considered less effective for NO_x reduction during biomass combustion. Also, the use of methane is not suitable because it increases the CO emission. Advantage of using ammonia is lower cost, compared to urea. On the other hand, urea is environmentally and health friendly, and its storage is easier. If urea is used, there is a possible increase in the harmful N₂O emissions [26].

2. Understanding of SNCR based on theoretical and experimental studies.

It is important to consider the most influential parameters for SNCR and the possibility of the appearance of by-products, such as N₂O, ammonia slip, ammonium sulfate salts, as well as the increase in CO emissions. The efficiency of the method is significantly influenced by the type of agent used, the amount and composition of the additive, the temperature, the residence time, the molar fraction of agent/NO, as well as the oxygen concentration in the flue gases [27].

The temperature range in which the SNCR method can be applied depends on the used reducing agent. For ammonia, the recommended temperature range is 1123-1273 K, and for urea is 1023-1323 K.

Table 1. Review of denitrification methods alongside with their main advantages and disadvantages

	Combustion control	SCR	SNCR	Wet scrubbing	Electron beam	Electrochemical method	Non-thermal plasma
Efficiency [%]	<80 ¹ [23]	60-90 [5]	30-70 [5]	96 ² [27]	82 ³ [28]	60 ⁴ [29]	14-96 ⁴ [27]
Temp. range [°C]	Optimal for combustion	150-590 [5]	800-1100 [5]	Ambient temp. [27]	90 ³ [28]	400 ⁴ [29]	200 ⁴ [27]
Advantages	Mature technology, Modification of the combustion process/ operating conditions on existing furnaces, without the use of additional pollution abatement equipment behind the main combustion zone	High efficiency, Good selectivity, Cost effectiveness, Relatively simple system, Mature technology	No waste generation, Simple solution, Easy implementation on existing combustion systems, Lower cost, Can be combined with other deNOx methods, Mature technology	Simultaneous deNOx and deSOx, PM and acid gases control, No high temperatures, No extraneous chemicals, Easy regeneration, Low cost of equipment and additives, Mature technology	Simultaneous deNOx and deSOx, No waste generation, By-product can be used for fertilizer production	Reductant is not required,	Simple system and operation, No waste generated, By-product are useful, Low cost of equipment
Disadvantages	Low efficiency, They are not enough for flue gas cleaning	High cost of catalyst, 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 temperatures, 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 in real-scale systems	Low efficiency, High energy consumption and cost, Operating pressure is low
Reference	[21,22,30]	[27,31-33]	[5,26,27,34]	[35,36]	[27,28]	[27,29]	[37,38]

¹depends on inlet NOx concentration, for industrial appliances high efficiency is not achievable so secondary measures are necessary

² for temperature 70°C with urea solution, lab experiment

³for simultaneous NOx and SOx reduction in lab experiment for liquid fuels

⁴lab experiments

Higher temperatures during the SNCR cause the decomposition of the reducing agent which leads to a decrease in the NOx reduction. If temperatures are below the recommended, NH₂ radicals do not react with NOx since the rate of reaction is slower. Also, during the reduction, the opposite effect from expected may occur if NH₂ radicals react with oxygen, which would result in the further NOx formation [5].

Residence time has a very significant impact on the NOx reduction when the SNCR method is applied. The time required depends on the available temperature range. The maximum reduction is achieved by complete mixing of the reducing agent

with the flue gases at the optimum temperature. The required residence time also depends on the flue gases velocity, their composition, as well as the construction of the furnace. The order of magnitude of the residence time for SNCR chemistry is a few seconds.

The implemented injection method affects the mixing effect. A normalized stoichiometric ratio (NSR) is an indicator of the injected optimal flow of the reducing agent. By optimizing the NSR, the maximum reduction is achieved, and the possibility of a part of the agent not reacting (emission into the atmosphere) is avoided (environmental impact and technology efficiency are minimized). To properly implement SNCR, it is necessary to take into account the choice of reducing agent injection site and the achievement of good mixing conditions. The effect of mixing is particularly significant at higher temperatures, at the upper limit of the SNCR range [28]. To overcome obstacles of nonuniform

distribution and poor mixing in industrial plants, injection is performed at several levels and in several places along the cross-section of the furnace (this is especially necessary for larger industrial furnaces, due to frequent temperature fluctuations in height and cross-section of the furnace) [31]).

The use of additives, i.e. enhancers can significantly contribute to NO_x reduction. In addition to the direct influence on the reduction efficiency, it can also lower the necessary temperature range for the SNCR application. The influence of additives is achieved due to the radicals they provide. This initiates chemical NO_x reduction reactions. Additives can reduce harmful flue gas components. To be properly applied, it is necessary to observe the temperature range in which the additives have the desired effect. At temperatures below the optimum, additional compounds may be formed which are undesirable. Besides, it can occur that part of the additive does not participate in the process and as a result is emitted into the atmosphere. On the other hand, at higher temperatures, very fast reactions not related to NO_x may happen. Additives of interest to SNCR are CO, H₂, CH₄, various combinations of hydrocarbons, alcohol additives (CH₃OH, C₂H₅OH), and sodium compounds (NaOH, Na₂CO₃) [29]. The presence of HCl can cause a decrease in the temperature range needed for SNCR, but its presence is only favorable at temperatures below 1173 K, because otherwise, it can cause an increase in NO formation [30].

3. Major challenges for SNCR application.

The most important challenge for SNCR implementation is to place the reducing agent where it is most effective in the furnace. The variations in operating conditions and furnace loads must be considered, meaning it must be injected in an appropriate temperature range, given that the temperature of flue gases varies both by cross-section and the furnace height. Viewed by cross-section, a higher amount of reducing agent must find its way to the furnace center where more NO_x can be found and compared to the cooler walls. Otherwise, NO in the center meets insufficient ammonia for reduction and excess NH₃ near the walls slips through (ammonia slip) affecting furnace's downstream equipment. Also, the flue gas temperature varies by height caused by changes in the furnace load or calorific value of the fuel (which is particularly important for biomass use). This may also result in increased deposits thickness on the heating surfaces, etc. At low-load operation optimum SNCR temperature region shifts within the lower part of the furnace, and *vice versa*. To overcome uneven temperature distribution and

imbalances from the furnace load changes, it is necessary to predict more reducing agent injection points by furnace's cross-section and levels. The presence of water has a positive effect on reducing NO_x emissions and direct injection into the flue gas stream can be performed. On the contrary, an increase in the H₂O concentration can lead to an increase in the NO content due to the higher conversion of NO to NO₂. Oxygen concentration has a significant effect on the possibility of NO_x reduction. When increasing the concentration of O₂ from 4% to 21%, at a temperature of 800°C there is a decrease in the NO_x reduction, because at high oxygen concentrations more ammonia is oxidized, which consequently leads to greater NO formation [31]. Simultaneous regulation of the emission of carbon monoxide, carbon dioxide, hydrocarbon compounds, and nitrogen oxides presents a great challenge. In SNCR carbon monoxide affects the NO reduction regime by shifting the temperature range for reduction to lower temperatures. In addition, there is a narrowing of the range in which it is possible to achieve a NO reduction, because of the increased amount of radical levels, the reverse process is created. Therefore, there is an increase in NO_x emission in the mixture CO₂/O₂, while for the mixture H₂O/O₂ there is a decrease. At lower CO concentrations, there is no disturbance in NO reduction [32]. Since both CO and NO_x are very unfavorable for the environment and health, it presents a great obstacle to reduce both of them at the same time. Nitrogen suboxide emission, N₂O, is expected to be low. According to [24], for 1000 ppm NO_x inlet concentration, due to the SNCR, the amount of nitrogen suboxide formed was 51 ppm when using ammonia, while for urea it was 157 ppm. The N₂O formation is mostly influenced by temperature, the amount of reducing agent injected, as well as the NO_x and CO concentration in flue gases. The formed N₂O does not have to be emitted completely, there is a possibility of its reduction to N₂. There is a possibility of combining SNCR with other measures. When stricter NO_x reduction is required, a hybrid SNCR-SCR system can be applied (SCR in the duct). When ammonia as a reducing agent is used, ammonia slip from SNCR is purposely generated to feed SCR catalyst. Reduction of NO_x to 90% with reduced SCR catalyst volume is possible with a hybrid system application [33].

4. Detailed chemical mechanism.

SNCR with ammonia as a agent [34] can often be found in the literature as thermal deNO_x, while SNCR in combination with urea appears as NO_xOUT [35]. When ammonia is used as an agent, its decomposition occurs, which generates NH₂

radicals that enter into a chemical reaction with NO, after which N₂ and H₂O are formed as products, while urea (NH₂)₂ is first decomposed into HNCO and NH₃. Afterward, in the case of urea, HNCO reacts with H and OH radicals, resulting in the formation of NH₂ and NCO, and only then, the radicals react with NO. Thermal decomposition of urea in real conditions does not occur instantaneously, so it is necessary to include the chemistry of this procedure in the chemical model of SNCR with urea. The selectivity of the process is determined as the ratio of reduced moles of NO_x and spent moles of reducing agent. The selectivity reaches a maximum when its value equal to one, while negative values of the selectivity indicate the reverse process from reduction [36]. Since the first appearance of this technology, a large number of studies have been presented, to explain the chemistry of reduction, as well as the conditions under which it happens. Table 2 shows the selected representative mechanisms, as well as the specifics that characterize them.

The importance of numerical simulations is emphasized as a guide for system design, due to the cost of experimental tests. To be able to numerically simulate SNCR, it is necessary to select the chemical equations that will represent SNCR. Otherwise, an overly large system of equations emerges which with conservation equations requires extensive resources besides other complications that arise. The selection of representative equations for SNCR chemistry is the key to making numerical modeling possible. There are several ways to perform this procedure. One is the systematic tabulation of chemical responses from canonical problems from combustion. These are solved with incorporated detailed chemistry for various parameterized initial conditions. The second approach is to implement

very simplified chemistry, i.e. a global reaction scheme. That implies limiting the reactions and species which will appear in the numerical simulation [37]. The second approach has proven to be very good for predicting process behavior. It is important to choose enough equations to simulate the SNCR reduction. In addition, not all chemical reactions contribute equally, so the global scheme, i.e. the skeletal reduction mechanism, can be distinguished from the detailed mechanism [38].

5. Numerical simulation and industrial application.

The reduction zone is characterized by a behavior very similar to plug flow behavior, preceded by a reducing agent injection zone in which conditions are different. Modeling droplets of reducing agent solution (mostly urea solution) during injection should be considered in real systems. This refers to the size and distribution of droplets. The injection is performed by spraying an agent which changes from a liquid to a gaseous phase and mixes with the flue gas, due to the temperature necessary for SNCR. So this is a multi-component problem. At temperatures higher than 673 K, the formation of a solid urea membrane around the droplet can be expected, in which water remains trapped in the initial moments of spraying [36, 43]. Also, it is necessary to pay attention to the very intense turbulent mixing in the injection zone.

NO_x emission from application in plants that use biomass as fuel with and without SNCR implemented is presented [2]. Authors showed that the emission of NO_x with implemented SNCR for mixed fuel in vibrating grate boilers is in the range of 0.343-0.384 kg/t, while without SNCR is almost three times higher.

Table 2. Detailed chemical models for SNCR

Author	Year	Reducing agent	Reactions	Species	Notes
Miller and Bowman [39]	1989	NH ₃	234	103	Thermal, prompt, fuel-N mechanisms, NNH
Coda-Zabetta and Hupa [40]	2008	NH ₃	371	60	Reaction subsets for oxidation of H ₂ , CO, Light hydrocarbons and CH ₃ OH alongside nitrogen pollutants
Javed <i>et al.</i> [41]	2008	Urea	170	30	Effect of CO and H ₂ addition
Klippenstein <i>et al.</i> [42]	2011	NH ₃	207	34	NNH mechanism for NO formation

CONCLUSION

The introduction of biomass, especially agricultural residues, into the energy sector represents a significant step forward in sustainable development. It is crucial to overcome accompanying problems and obstacles. Here, the SNCR is considered as a solution for one of the negative effects of biomass combustion - NO_x emissions. The NO_x emission contributes to further damage to the ozone layer and is harmful to human health and the entire ecosystem. This paper presents the concise view of the SNCR, from the initial theory, to the possibility of implementation in a real-scale systems. It is necessary to further expand the knowledge about this measure, especially to overcome the lack of data on the application of SNCR in the energy sector. In addition, there is a need for further research on the numerical simulation of the SNCR process, especially when urea is used as a reducing agent. Overcoming these limitations is the aim of the future research, so the optimal level of knowledge can be accomplished.

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NOMENCLATURE

SCR - selective catalytic reduction;
 SNCR - selective non-catalytic reduction;
 NO_x - nitrogen oxides (NO+NO₂).

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