

Comparison of NO_x Emissions Decreasing Methods for Biofuel Boilers

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Abstract. The main idea of research is to figure out the emissions of nitrogen oxides reduction using various type of reduction methods. In experiments were used NO_x reduction methods: high CO emissions generation, flue gas recirculation, water and water vapor supply, selective non-catalytic reduction (SNCR), and SNCR with flammable additive. This study presents emission and combustion results obtained burning furniture production waste which generates higher rate of NO_x emissions. The result of research shows, that CO emission has the biggest impact factor -on reducing NO_x emission. Burning fuel in combustion zone with first and secondary air ratio (40/60) and using methods for higher generation CO emissions reached 3.000 mg/m³ which reduces NO_x emissions up to 83%. Using selective non-catalytic reduction with traditional and flammable additives reduction of NO_x emissions reached up to 55%.

Keywords: biomass, combustion, NO_x.

Conference topic: Environmental protection.

Introduction

This work was aimed at a variety of methods to reduce nitrogen oxides while burning pellets rich of nitrogen in their mass in a moving grate boiler. Bigger nitrogen content in fuel mass which in combustion process it generates more nitrogen oxides. In thermochemical reactions nitrogen splits out from the fuel and combines with oxygen contained in the combustion chamber thus allowing to form NO and NO₂ compounds, generally naming them NO_x. Fuel nitrogen oxides emissions can be reduced by flue gas cleaning or by influencing the combustion process directly (Wang 2003).

Were used method for primary NO_x reduction: fuel blending, boiler power decrease, a small amount of excess air, air distribution or flue gas recirculation to the combustion chamber and secondary NO_x reduction measures: selective catalytic reduction (SCN) and selective non-catalytic reduction (SNCR).

This study presents an experimental investigation of various type of NO_x reduction methods in the temperature range of 1000–1200 °C. Basic fuel used in the studies is known for higher nitrogen content. Up to 1400 mg/m³ NO_x concentrations were measured in the flue gas while burning furniture production waste pellets.

This work assesses the influence of NO_x reduction by various type of NO_x technology. NO_x concentrations were measured during this research and the effect of each method was compared. NO_x reduction while burning furniture production waste showed that primary NO_x reduction methods can reduce NO_x by 83%. Using selective non-catalytic reduction with traditional and flammable additives reduction of NO_x emissions reached up to 55%. Results showed that NO_x emissions can be rapidly decreased by combustion properties regulation and this technology can be easily installed in the existing boilers.

Method

Experiments were carried out in the Combustion research laboratory of Kaunas University of Technology, which has an installed pellets-fired industrial boiler model and all other system elements.

The main component of the stand is furnace (Fig. 1) which has systems for fuel and air feeding. Furnace and heat exchange surfaces, which are installed above the furnace are the main two components of the water boiler. The design capacity of the boiler is up to ~50 kW. To keep stable pressure in the combustion chamber, draft fan with a frequency converter assists to emit flue gases to the chimney.

This boiler has all the technology elements which allow imitate industrial boiler with moving grate. Boiler can be regulated by firstly and secondary air inlet, it can be adjusted at constant suction pressure and constant temperature of boiler. Set temperature is supported by water circulation section with heat exchanger elements.

There are two parameters that control combustion of all used fuel types, first is fuel feeding rate and second – the amount of air that is supplied to the combustion chamber. Generated pollutant concentrations are measured by flue gas

analyzer. This analyzer measures temperatures and concentrations of nitrogen oxides, nitrogen monoxide, oxygen and carbon monoxide in the exhaust gases.

Frequency converters, which are mounted on the side of the furnace, are used to determine and regulate the constant rate of fuel feeding and the amount of air for combustion.

Material

The experiments were accomplished on basic fuel – furniture production waste. Furniture production waste was used as high NOx concentration generated fuel (with high nitrogen content in mass) and dry fuel. Table 1 illustrates results of the used fuel composition and calorific value.

Table 1. Properties of the substances used in the combustion research

	Fuel moisture content W, %	Carbon content, %	Nitrogen content, %	Hydrogen content, %	Sulphur content, %	Lower calorific value $Q_{w.f.1}$ kJ/kg
Furniture production waste	4.9	48.74	3.47	5.7	<0.01	17 900
Wood pallets	8.7	54.71	0.2	3.0	<0.01	17 100

Furniture production waste is formed in the furniture production companies. During the technological process of furniture production some unused parts of wood chipboard, sawdust and defective products remains as a waste. Wood is not the only substance that this waste consists of, materials such as chemical substances used for treating wood, furniture laminate, cardboard and residues of other packaging can be also found in this waste. Consequently, wood processing companies want to use this waste to meet their own energy needs.

In Lithuania, there are a lot of companies that produce large amount of such waste and the problem is that this waste cannot be used in energy production, because during combustion process it produces high concentrations of NOx. However, successful reduction of nitrogen oxides, which are formed from fuel bound nitrogen, could mean that furniture production waste fuel can be used in energy production. This would also benefit the environment in the way that there would be less disposed waste and it would also decrease the amount of other biofuel required for energy production.

Experimental fuel combustion research

Experimental fuel combustion research was performed with the aim to examine the efficiency of lowering measure of NOx in flue gases. The fuel was burnt so that the temperature in the combustion zone will not exceed 1200 C°; burning at such temperature mode would allow examining solely the generation of “fuel NOx”, because this temperature is insufficient for the formation of “thermal NOx” (Hodžic *et al.* 2016). At this temperature range its share in the NOx content is so small, that it has no practical value.

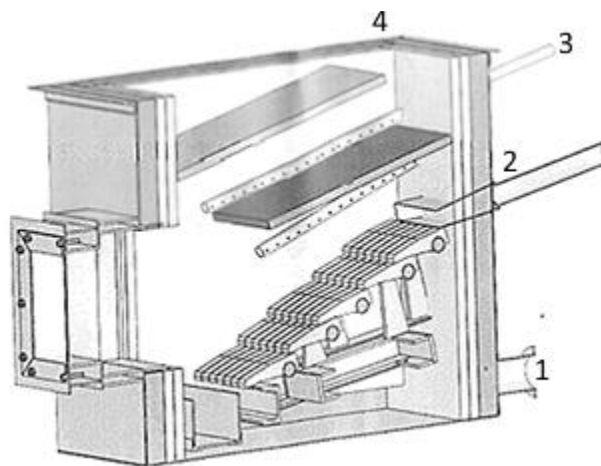


Fig. 1. Furnace: 1 primary air; 2 fuel inlet 3 secondary air and additives supply; 4 exhaust gasses

Before the nitrogen oxides reduction studies was measured the compound of exhaust gases while burning furniture production waste. Nitrogen oxides at a 40/60 ratio of air are determined, at the constant power of 36 kW and at sufficient combustion, which describes the amount of CO in the flue gases. Fig. 2 presents the main CO and NOx concentrations in flue gases dependence of excess air factor.

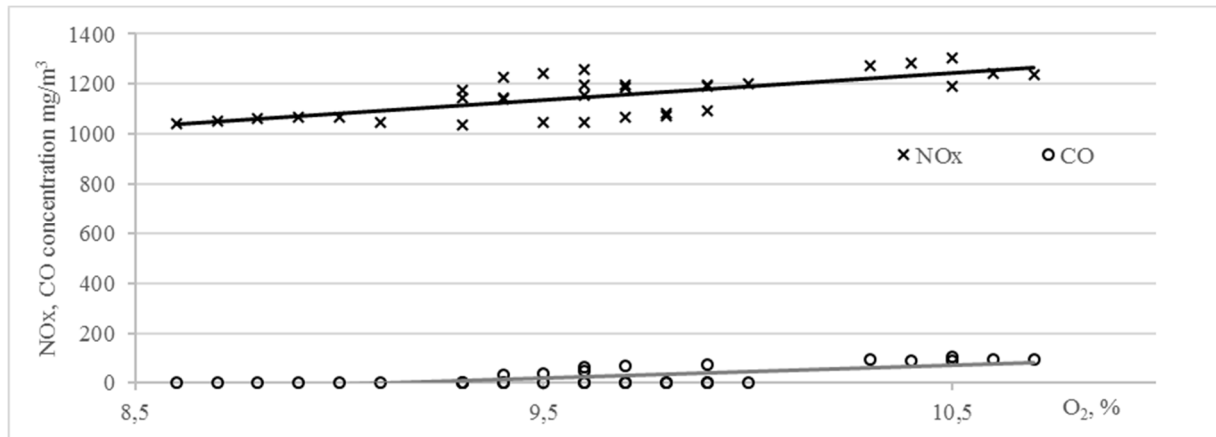


Fig. 2. NOx and CO values in furniture production waste combustion depending on excess air

Nitrogen oxides concentration increases with air excess in the flue gases. Lowest NOx concentrations 1108 mg/m³ recorded at 8,6 % excess air. The highest recorded NOx concentration totaled 1303 mg/m³ when 10,7 % excess air was achieved while burning furniture production waste, with nitrogen content in the substance accounting for 3.47 %. This shows that big amount of N content in the substances turns to nitrogen oxides.

Primary NOx reduction method

Initial concentrations of nitrogen oxides reduction methods employs as a preventive measures to reduce NOx. Their operating principle is based on the reduction of oxides of nitrogen in their formative stage. NOx reduction performed through the predefined furnace construction: air / fuel supply place in the furnace, quantity of primary/secondary air, fuel composition. These measures, during boiler operation time, does not require significant energy and financial resources.

Fig. 3 shows the graphical results and they will be summarized in the following chapters.

CO generation effect

The combustion air is supplied into the two combustion zones (primary and secondary) separately. This design allows to adjust the total amount of oxygen in the flue gas and primary / secondary air ratio. It is also possible to obtain different combustion modes. Without the supply of air to the secondary combustion zone the chemical reaction time is extended. The carbon monoxide and nitrogen oxides take place in competition with the reaction – the conversion of the nitrogen oxides to N₂, oxygen is used for oxidizing CO to CO₂.

Oxygen content in flue gas was varied between 4% and 10%. At low excess of oxygen, the oxygen content in the flue gas is 4%, inside the furnace, in the primary combustion zone very high CO is generated as much as 2,900 mg/m³. At this level, the NOx decreased to 200 mg/m³. When further reductions of carbon monoxide is performed by increasing the supply of air, nitrogen oxides concentration increases until it reaches the combustion usual concentration.

Flue gas recirculation effect

By reducing the amount of oxygen in the primary air in the flow from 21% to 13%, there was a no significantly influence to the NOx concentration. The supply of a larger proportion of recirculating flue gas, nitrogen oxides concentrations slightly increased. Combustion temperature does not have any significant effect for the formation of fuel NOx. The concentration of oxygen in the combustion zone, where all of combustion air is supplied through the grate, in any case, is much higher than the stoichiometric air-fuel ratio. Locally, in the fuel combustion layer, the excess air is much higher than 1, because only part of the air is consumed in the combustion of fuel layer. Grater amount of air is used for gaseous gasification products combustion in the zone above the grate.

Recirculating flue gas supply to the secondary air channels had a slight impact on the increase in the concentration of nitrogen oxides. By reducing the oxygen content in the secondary air from 21% to 13% increased the NOx concentration from 450 mg/m³ to 550 mg/m³. Recirculating flue gas supply into the secondary combustion zone does not have a significant effect the concentrations of NOx reduction.

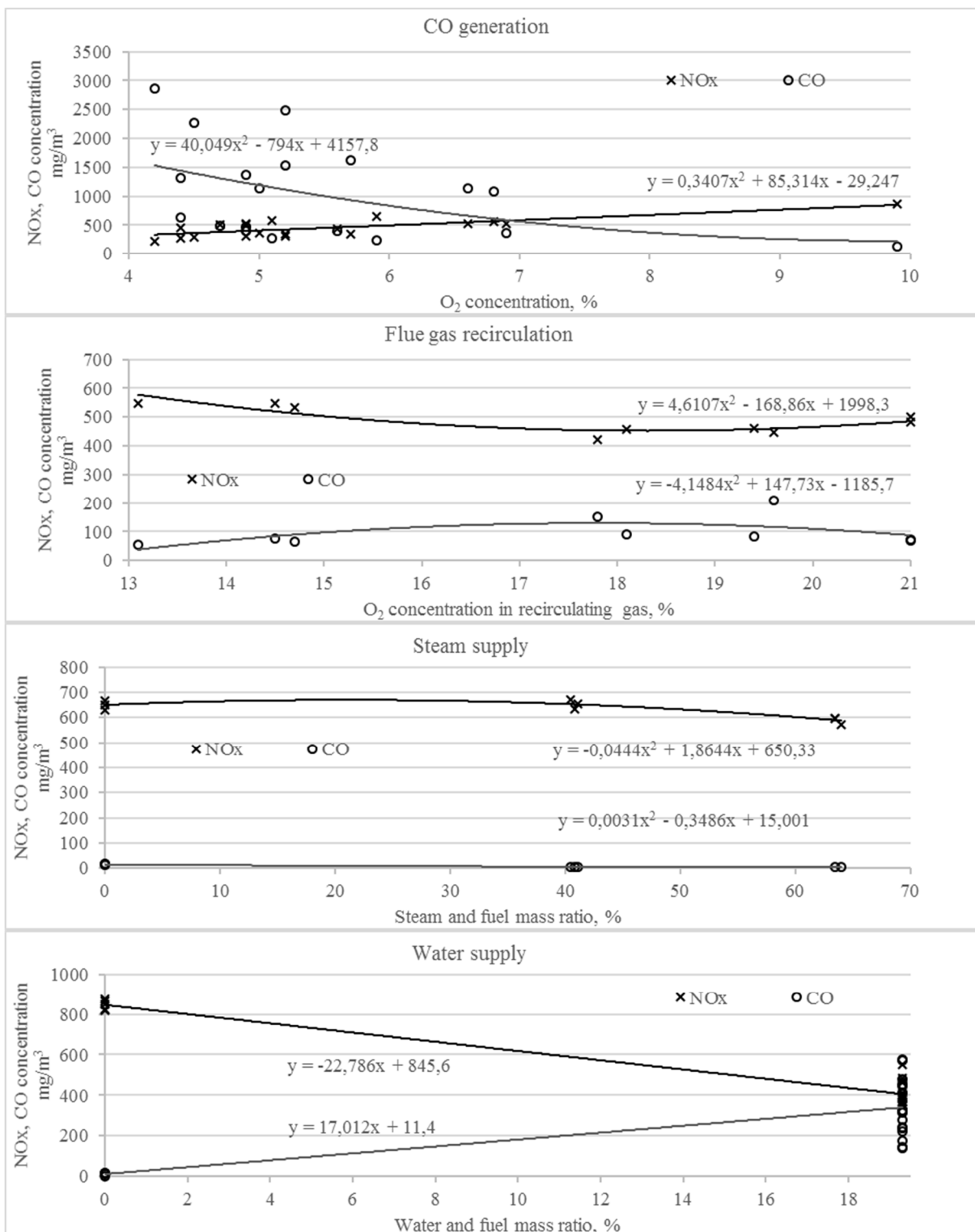


Fig. 3. NOx and CO values by primary NOx reduction method

Steam supply effect

Steam was supplied to the combustion zone above the grate. In contrast to the supply of room temperature water, steam has a high energy content. As a result of the experiment there was no flue gas temperature decrease. Because of the high steam energy potential, furnace cooling and carbon monoxide formation has been avoided.

When the amount of steam supplied was 40% of the fuel mass, it did not have a significantly influence the NOx concentration. The low NOx decrease can be explained by the decrease of oxygen concentration in the combustion

zone, where water evaporates and act like a balast in the combustion zone. When this amount of steam generates nitrogen oxides concentration reduces from 850 mg/m³ to 820 mg/m³.

The maximum NOx reduction efficiency in this study was achieved when the amount of steam supplied was 65% of the fuel mass. NOx concentrations were reduced from 850 mg/m³ to 750 mg/m³. Supplied steam had a marginal impact on the reduction of nitrogen oxides.

Water supply effect

Secondary air channels have been selected for the water injection. Due to the high temperature of 1350 K in the secondary combustion zone, the water evaporates and mixes with the secondary air. For water evaporation, depending on the quantity of water, certain amount of thermal energy is consumed. For this reason, the area to which the water is fed, is cooled. Combustion reaction rate has a direct dependence on the temperature. As the temperature decreases from 1320 K to 1120 K incomplete combustion products CO and C_nH_m concentration increases. Increased concentration of carbon monoxide leads then to conversion with the nitrogen oxides. During this conversion of CO reduces the NOx concentration.

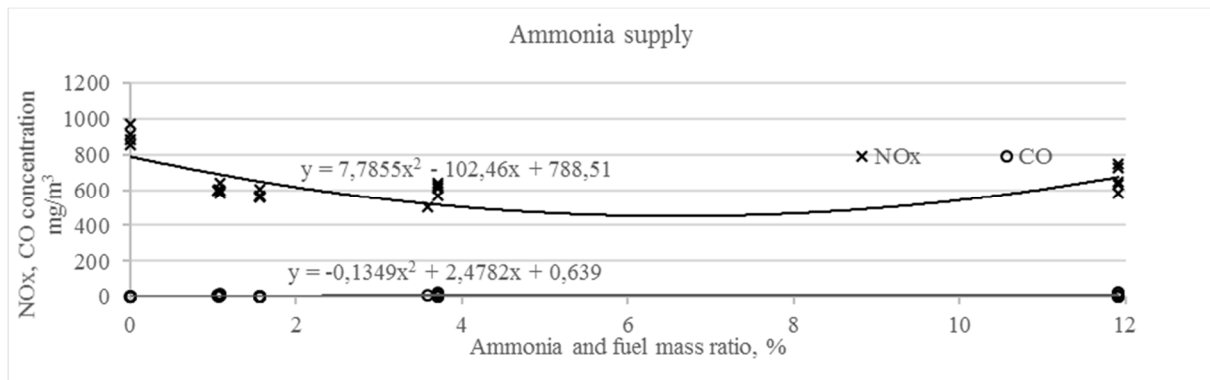
When the amount of water supplied was 19% of the fuel mass, CO concentration increased on average to 340 mg/m³. NOx concentration was reduced twice, on average to 450 mg/m³. Such a significant reduction in NOx may be due to local oxygen concentrations decrease in the combustion zone due to the high steam content. Also flame temperature decreased. These factors influenced the small combustion quality deterioration, causing a generation of incomplete combustion products like CO and C_nH_m. This has led to competition between the oxidation of CO and N in situations that allows to decrease NOx concentrations. Another reason why NOx reduces may be due to reduction in the O₂ concentration in the reaction zone, which directly affects the fuel NOx formation.

Secondary NOx reduction method

Secondary measures, often referred to as the “end of pipe” means. These are the measures which require complex and expensive equipment (SCR, SNCR methods) and chemical reagents (NH₃, NH₄CO and other). These measures require considerable financial costs. You need to maintain periodic equipment maintenance, perform maintenance, buy chemicals. However, it ensures a stable and desired NOx reduction effect. Experimental stand has been equipped with additional equipment for experiments using SNCR technology.

SNCR reaction is effective in a temperature range of 900–1100 °C called temperature window. The temperature window is so narrow that ammoniac reagents could not mix with flue gas thoroughly and have no enough residence time to decompose and react with NOx adequately, which leads to lower NOx reduction efficiency (30–50%) (Tayyeb Javed 2007).

Below are the graphical results and they will be summarized in the following chapters.



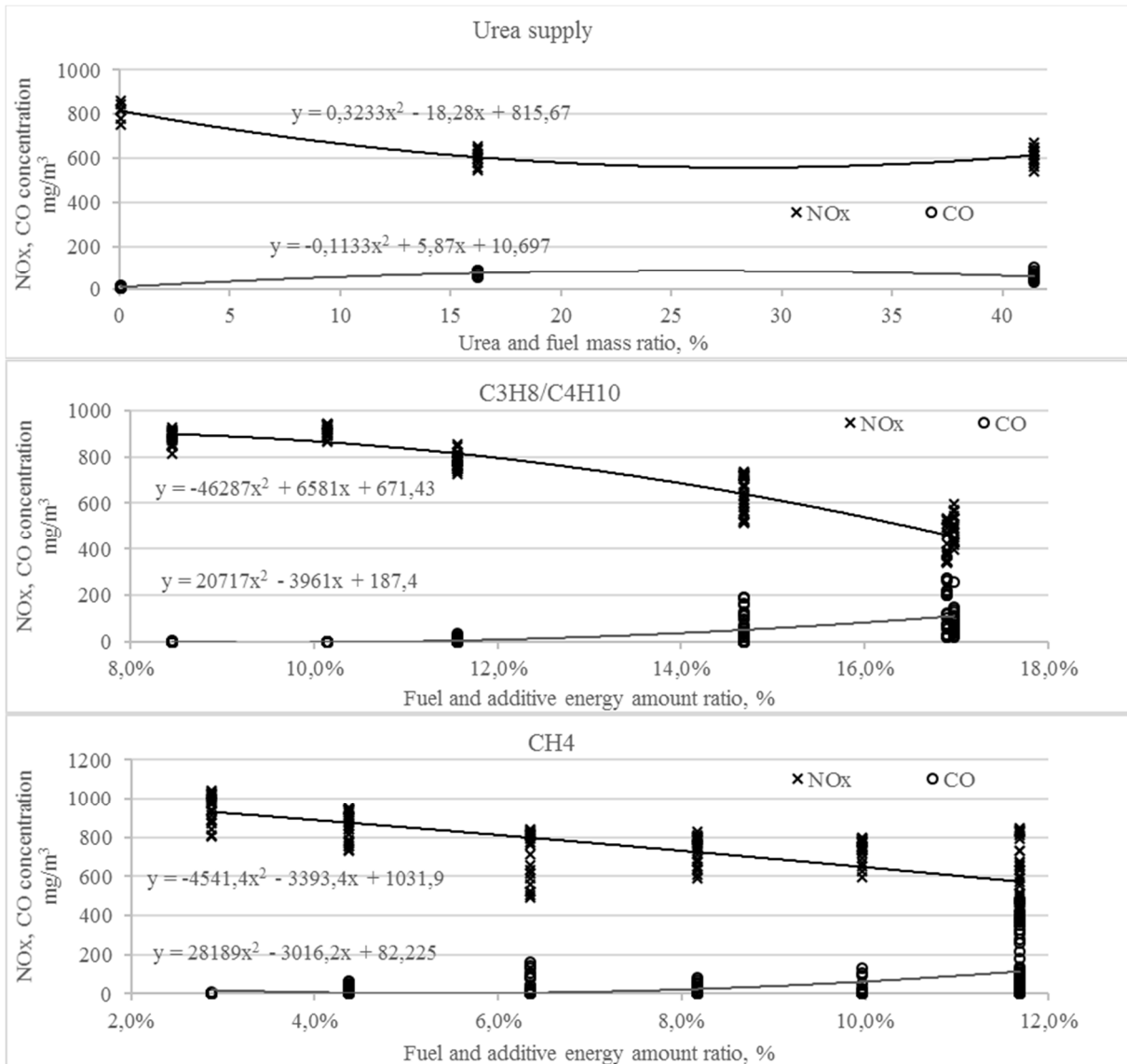


Fig. 4. NOx and CO values by secondary NOx reduction methods

Ammonia supply effect

Ammonia water 10% solution has been prepared for the research. During the experiment, the dropper was set to supply amount of solution through the secondary air channels. Ammonia water drip points were set at 1.1%, 1.6%, 3.7%, 11.9% of the fuel mass. After each phase of the experiment, the infusion of different quantities of ammonia water, some time past due to the boiler operating mode and the emission concentrations settling. The supply of any quantity of the solution into the secondary combustion zone was observed sharp decrease of nitrogen oxides.

The maximum nitrogen oxides reduction efficiency was achieved by injecting 1%–3.7% ammonia solution of the fuel mass. By increasing the amount of ammonia, higher reduction efficiency has not been reached. This may be due to the fact that at the injection point, all ammonia is used for NOx decomposition reactions and NOx disassembling was fully completed in this zone. However, part of the flue gas flow could not enter the ammonia injection zone and pass without effect. Further increasing the amount of ammonia had no impact. We can guess that, in order to increase ammonia impact on the NOx we should not only increase the amount of this solution, but also the injection site or split the ammonia flow to multiple injection sites, in order to cover potentially higher flue gas flow.

Urea supply effect

Of total urea nitrogen mass fraction is 46.5%. Prepared solution of urea concentration is 9%. As well as the dosage of ammonia during the experiment the amount of solution was established in the dispenser. It was 16.2% and 41.4% of the fuel mass. After each experiment stage of infusing different amounts of ammonia water, some time past due to the

boiler operating mode and the emission concentrations settling. When the solution was supplied to the secondary combustion zone it was observed sharp decrease in nitrogen oxides.

NOx concentration in the flue gas, converted to 6% oxygen, averaged to 1010 mg/m³. When supplying 16% of the fuel mass amount of solution NOx concentration decreased to an average of 705 mg/m³. When supplying higher amount, 41% of the fuel mass amount of solution, NOx concentration decreased to an average of 680 mg/m³. Increasing the supply of urea solution, the concentration of CO increased to 70 mg/m³.

C₃H₈/C₄H₁₀ supply effect

Flammable additives (hydrocarbons) are chosen to extend the reaction time between CO and NO and temperature range (Zhang *et al.* 2008). The propane-butane mixture was supplied to the secondary ducts with a secondary air. Given the other, previous experimental findings it has been chosen to supply from 130 up to 300 l/h flow rate. When the gas meter volume of gas delivered within the stipulated time was set, gas flow was calculated. Assuming use of propane-butane is 1:1 on the basis of the weight of 1 m³ of gas under normal conditions, the feed gas weight was calculated. Once the supply of propane-butane gas was initiated, a significant NOx reduction was obtained (Fig. 4). Boiler thermal power gain was also observed. At the beginning of the experiment boiler thermal input was 17.3 kW, and when gas was supplied – 21.0 kW. This higher power gain is explained by additionally using a more than doubled propane-butane gas calorific value compared to the furniture industry waste pellets. The supply of gas decreased oxygen excess.

When the amount of propane-butane gas mixture supplied was 17% of the fuel mass, NOx was reduced from an average from 960 mg/m³ to 450 mg/m³. This decrease could have been because of the active hydrocarbons supply into the secondary combustion zone. Active hydrocarbons splits into lighter intermediate hydrocarbons including CH₄, which reaction with NOx pushes it to dissociate into elemental nitrogen and diatomic oxygen. Carbon monoxide concentration increased slightly – from 0 to 100 mg/m³ in all gas supply range.

CH₄ supply effect

The methane was supplied to the secondary ducts with a secondary air from 130 up to 560 l/h flow rate of CH₄. As figure out that in the absence of free oxygen content, nitrogen secluded from the solid fuel can hardly form a compound with oxygen to NO and NO₂. Therefore, while the amount of excess air is decreasing, decreased free oxygen content is one of the reasons why NOx concentration in the flue gases is reduced (Plečkaitienė *et al.* 2013; Carroll *et al.* 2015).

When supplies of CH₄ reached 117% of the fuel mass, NOx was reduced from an average –1010 mg/m³ to 390 mg/m³. Carbon monoxide reacting the same as supplying mixture of propane butane.

Comparison of NOx reduction efficiency

To compare different methods of NOx reduction is difficult to it right because these methods are reacting not the same in different boilers and other conditions. Comparison of NOx reduction efficiency was made by the results which are obtained during these research shown in Table 2.

Table 2. Comparison of NOx reduction efficiency

NOx reduction method	Primary NOx value	Reached NOx value	NOx reduction efficiency
CO generation	950	200	79%
Flue gas recirculation	520	500	4%
Steam supply	850	750	12%
Water supply	900	450	50%
Ammonia supply	900	570	37%
Urea supply	1010	680	33%
C ₃ H ₈ /C ₄ H ₁₀ supply	960	450	53%
CH ₄ supply	1010	390	61%

The main results show that the biggest impact factor for NOx reduction has CO generation method. Other promising methods are water supply and SNCR technology with flammable additives.

While using flammable additives we observed not only significant decrease of O₂ in excess air (Fig. 4), but also increase of CO. Unlike using conventional NOx reduction additives where CO remain the same in all the additives supplied range (Kuang 2014; Wang *et al.* 2012). That directs that the main effect of NOx reduction is CO gasses.

Conclusions

- Generation of a large CO concentrations in combustion zone technology has a highest impact to NOx reduction. With the biggest CO concentration – 2900 mg/m³ NOx value decreases to 200 mg/m³. Which is more than 70% reduction efficiency. This method can be easily adopted to existing boilers and let them meet new environmental protection directive.
- Water supply and SNCR technology with flammable additives shows more than 50% NOx reduction efficiency. These technologies can be used together with CO generation method to ensure the best reduction effect.
- Ammonia additives has a high efficiency in small quantities (up to 6% of the fuel mass). Flammable additives at small quantities are less efficient, but when supplied more than 8%, NOx reduction effectiveness outperforms ammonia additives. Flammable additive act effectively in wider supply range from 3% to 10% results in NOx concentrations reduction from 900–1100 mg/m³ to 390–450 mg/m³.

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