

THE FACTORS WHICH HAVE INFLUENCE ON NITROGEN CONVERSION FORMATION

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Abstract. The European Union's normal proposal - to use 12 percent of indigenous fuel sources in Lithuania – make good conditions for expanded use of wood waste in all balance of energy. Biofuel use greener due to the fact that burning the carbon dioxide depends on natural carbon cycle flow and does not increase greenhouse effect, and does not include the sulfur dioxide. However, the ecological importance and combustion products resulting from nitrogen oxides and carbon monoxide. In the view of nitric oxide formation and the negative impact on the environment (Buinevičius and Puida 2005), it is necessary to reduce their concentrations in the atmosphere with emissions from fuel combustion in smoke. The aim of this work - using biofuels and wastes in the experimental studies, to identify the factors which have influence on nitrogen conversion coefficient formation. The paper presents selected fuels combustion technique, the measured concentrations in smoke are compared with the permissible concentration limits, as well submit as the nitrogen conversion into nitrogen oxides the dependence of nitrogen content in the fuel and oxygen content supply to the furnace. In addition, was additionally tested with varying quantities of urea-water solution sprayed pellets. It was found that the coefficient of conversion decreases with increasing nitrogen content in the incinerated fuels, and vice versa - with increasing oxygen content in the furnace, increasing the nitrogen conversion to nitrogen oxides. Also, was found influence of fuel density to NO_x formation, submit calculation methodology of the nitrogen conversion coefficient.

Keywords: nitrogen oxides, biomass combustion, conversion, conversion factor, NO_x concentration, wood pellets, carbamide.

1. Introduction

Lately more attention has been paid to biomass combustion. Ecological properties of biofuel are predetermined by the fact that carbon dioxide, generated during combustion, depends on natural carbon circulation cycle flow and does not increase greenhouse effect (Wiedlich 1991), and sulfur dioxide is not generated (Lakasavičius *et al.* 1997). This compound has an impact to rainwater oxidation (Verein 1983) and stipulates all hydrospheric, floral and material valuables impact (Benecke 1987; Smith 1981; Gauri and Holdren 1981).

The main contamination sources of man-made environment include: manufacture of products, transport, intensive cattle breeding. The major part of contamination – about 90%- is generated during combustion process: thermal power plants, industrial and domestic boiler plants, vehicle engines, etc. (Baltrėnas and Zagorskis 2008).

Nitrogen oxide is one of the main contaminants of the atmosphere. It can be generated in the following cases:

- “thermal”, molecule breakdown into atoms and radicals is supported, depends on temperature;

- “rapid“, takes place at the beginning on torch, radicals CH, CH₂ are involved, low-dependent on temperature and high-dependent of fuel molecular structure (fuel type);
- “fuel”, depends on nitrogen content in fuel and excess air factor.

Generation of nitrogen oxide is sufficiently thoroughly considered on the basis of chemical combustion reaction (Hill and Smoot 2000; Bowman 1998; Miller *et al.* 1998; Smoot *et al.* 1998).

In order to reduce NO_x, released with smoke to the environment, most scientists investigate and analyze reduction of nitrogen oxide concentration via various techniques (Buinevičius and Puida 2005, 1999, 1998; Puida 1998; Lapienienė and Šležas 2002; Buinevičius and Strakšys 2002; Melkūnas and Buinevičius 2007).

For example, the method of nitrogen oxide reduction during gas combustion process by off-stoichiometric combustion, i.e. zoning of gas combustion, dividing the required air or gas flow into two parts (Puida 1998). This method succeeded in NO_x concentration reduction to 35%, all fuel combustion conditions were observed. The investi-

gations for the purpose of NO_x concentration reduction by injection of selective reagents (Buinevičius and Puida 2005; Plečkaitienė 2009) were performed. This method succeeded in NO_x concentration reduction to 40%.

While searching for other fuel alternatives – complex fuel – meat and bone powder- was investigated (Buinevičius 2009, 2008; Buinevičius and Kazilevičius 2004). It was determined that only about 0.8 % of fuel nitrogen converts into nitrogen oxides. NO_x and CO concentration are very dependent on temperature level in combustor, fuel supply and grid driving mechanism operation.

All these shows huge interest in NO_x concentration reduction, however, during active use of renewable energy source it is necessary to find additional methods for NO_x reduction.

Therefore, the work objective is to investigate nitrogen oxide concentration in end products of biofuel and waste combustion and determine factors affecting nitrogen conversion coefficient formation.

2. Technique

Investigations were performed using biomass combustion stand from Kaunas University of Technology (KUT) Heat and Nuclear Power Engineering Department (Fig 1).

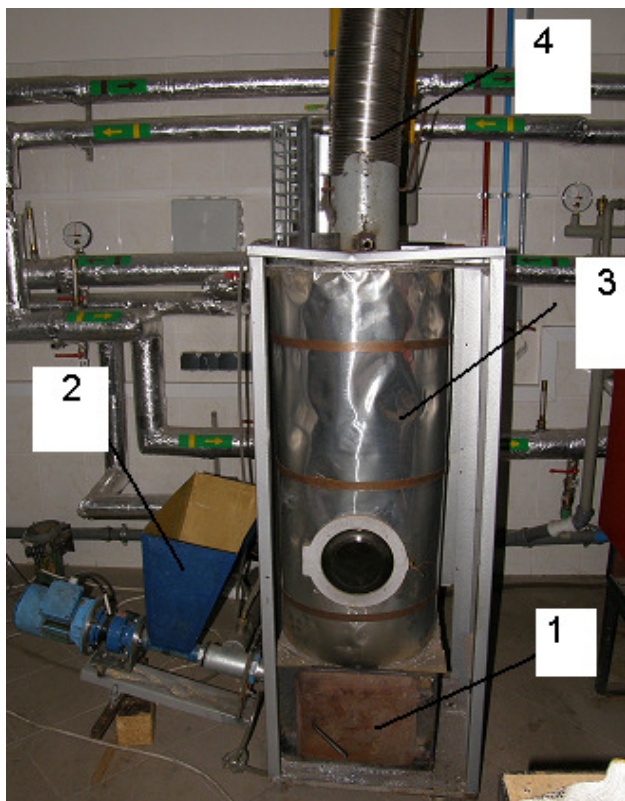


Fig 1. Stand of biomass combustion: 1 – combustor; 2 – fuel capacity; 3 – water boiler; 4 – chimney

Biomass combustion stand includes combustor 1 with fuel and air feeders. Boiler 3 is installed above the combustor, combustion products are removed to the chimney 4. An outlet for smoke analysis and temperature measure-

ments is installed in the chimney. Draft is natural it is controlled by valve which is installed in a boiler. Burner is installed in a combustor where fuel from fuel bin is supplied to grids with the help of screw. Air to the burner is supplied by a fan and is divided into two flows – one flows under the grids, another – above the fuel bed. During investigations the temperature and content of end product of fuel combustion was measured by gas analyzers.

The analysis of nitrogen contained in investigated materials was performed in LAI Agrochemical Research Center and in the above-mentioned KUT Department (Table 1).

Table 1. Characteristics of investigated fuel types

Fuel type	N content, %	Heat generation, kJ/kg	Ash content, %
Wood pellets	0.13	17537	0.171
Corn stalk pellets	0.590	15617	2.7
Buckwheat hull	0.574	15912	1.45
Cornstarch pellets	1.235	14934	0.95
Shredded packs (tetrapacks)	0.021	19451	13.13
Scrap wood and wood pellets	0.217	16333	1.36
Straw (80%) and fuel oil (20%) pellets	0.440	15743	8.47
Lignin (80%) and fuel oil (20%) pellets	0.870	18182	15.08
Mixture of all fuel types (fuel oil, straw, diesel fuel, etc.)	0.370	17024	10.22
Wood pellets with carbamide	0.3	17282	0.280
Wood pellets with carbamide	0.5	17423	0.295
Wood pellets with carbamide	1.0	17508	0.367
Wood pellets with carbamide	2.5	17582	0.438

Accumulation of physical and chemical processes begins in fuel bed. These processes depend on initial conditions: fuel, supplied air amount, its temperature, etc. Experimental investigation (Kavaliauskas *et al.* 2003) of impact of initial conditions to combustion process characteristics (combustion front velocity, its temperature and concentration of end product of combustion, such as CO, CO₂ and NO) were performed in fuel bed. It was observed that fuel density is one of the most important parameters for NO generation during combustion process, using biomass fuel of different type.

Current nitrogen content also has impact to NO_x concentration in end combustion products. Nitrogen conversion into nitrogen oxides K_N is calculated as the part of nitrogen contained in fuel converted into nitrogen monoxide. General equations are used for calculation of conversion ratio K_N (Buinevičius and Kazilevičius 2004).

Theoretical air:

$$V_o = 0,00027 \cdot Q_z + 0,0234, \quad (1)$$

$$V_d = 0,00025 \cdot Q_z + 0,9756, \quad (2)$$

here: V_o – theoretical air, m^3/kg ; V_d – theoretical combustion product volume, m^3/kg ; Q_z – the lowest value of heat generated by fuel, kJ/kg . This is considered the most important wood fuel energy parameter.

Ratio of nitrogen conversion into nitrogen oxides is calculated as follows (Buinevičius 2009):

$$K_N = \frac{(V_d + (\alpha - 1) \cdot V_o) \cdot C_{NO_x}}{328,6 \cdot N_K}, \quad (3)$$

here: K_N – fuel nitrogen conversion ratio, percentage; α – coefficient of excess air contained in combustion products; C_{NO_x} – concentration of nitrogen oxides in combustion products, mg/m^3 ; N_K – nitrogen content in fuel, percentage; 328,6 – conversion factor.

According to the European Union Directive contamination generated in end products of fuel combustion should not exceed permissible values for combusted fuel. For contamination generated during combustion of complex pellets different regulations for waste, solid fuel or biofuel are applied (Table 2).

Table 2. Concentration rates for biofuel and waste combustion

Fuel	Biofuel <1 MW	Waste
NOx, mg/m^3	750	400
CO, mg/m^3	4000	50

3. The Results

Values of NOx and CO measured during experiments did not exceed the limit of biomass fuel combustion product release permissible in Lithuania. Minimum CO concentrations were measured during combustion of scrap wood and wood pellets.

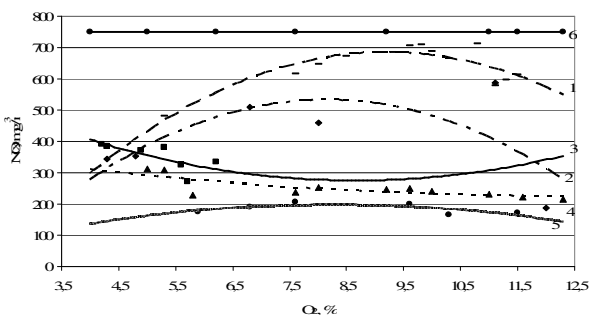


Fig 2. NOx dependence from oxygen during biomass fuel combustion: 1 – cornstarch pellets; 2 – corn stalk pellets; 3 – buckwheat hull; 4 – scrap wood and wood pellets; 5 – wood pellets; 6 – concentration rates for NOx

Wood pellets are well combusted (Fig. 3). Maximum measured NOx value – 205 mg/m^3 . Combustion quality can be seen from the diagram.

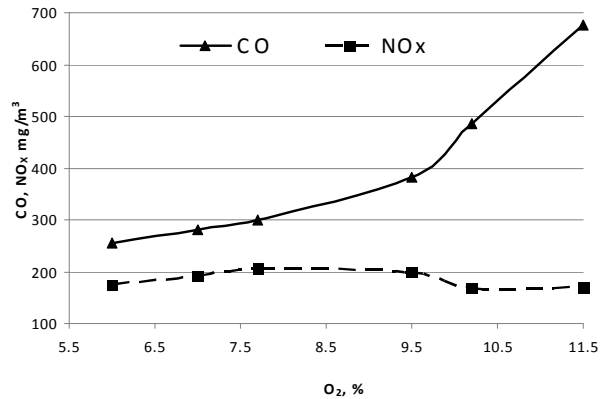


Fig 3. NOx dependence from oxygen during wood pellets combustion

Combustion of scrap wood and wood pellets (Fig. 4) was the same as combustion of wood pellets. Fuel is clean, oxide concentration are low.

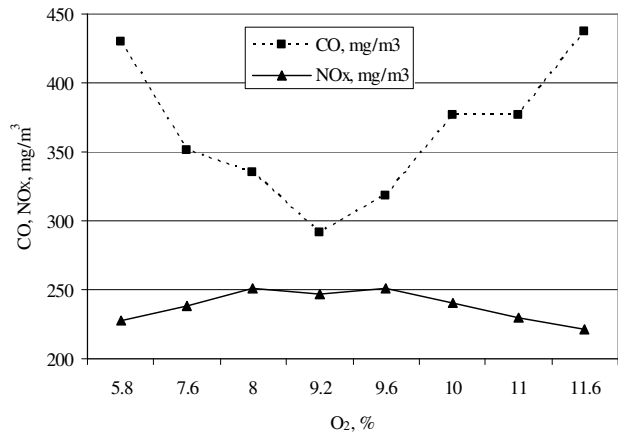


Fig 4. NOx dependence from oxygen during scrap wood and wood pellets combustion

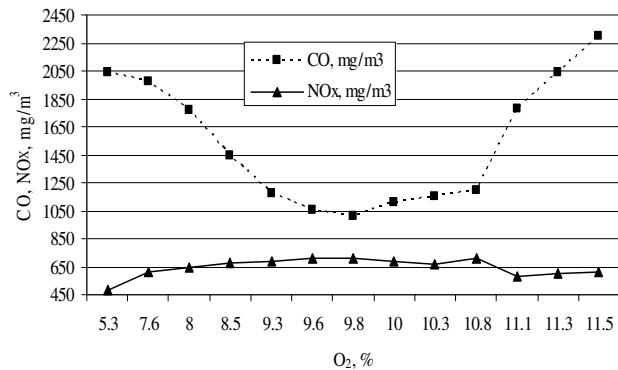


Fig 5. NOx dependence from oxygen during cornstarch pellets combustion

Cornstarch pellets combustion is difficult, combustion takes place only pushing off fuel bed and further-

more, there is a lot of burnout. Maximum measured NOx value – 713 mg/m³ (Fig 5).

Buckwheat hull is well combusted, combustion is complete, little ash; however the fuel has simple structure and it is difficult to optimize air in smoke duct (Fig 6). When air velocity increases, mechanical blow-off begins. So, it is difficult to optimize amount of air, supplied to smoke duct. During testing minimum oxygen concentration 4 % was not reached. Maximum measured NOx value – 392 mg/m³.

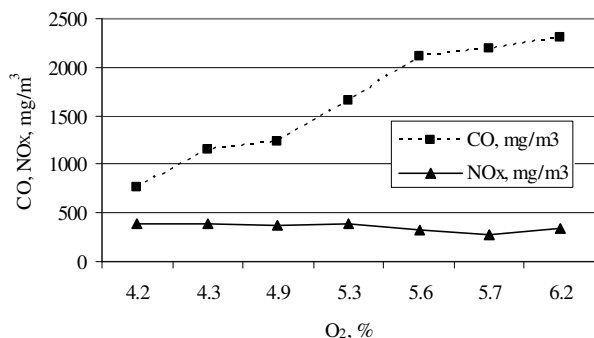


Fig 6. NOx dependence from oxygen during buckwheat hull combustion

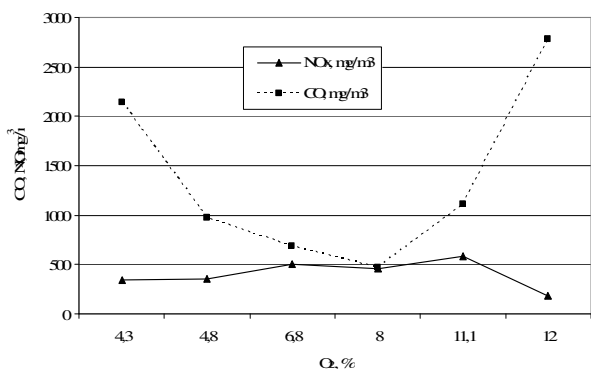


Fig 7. NOx and CO dependence from oxygen during corn stalk pellets combustion

Combustion of cornstarch pellets is similar to combustion of wood pellets, although after combustion of cornstarch pellets there are more of nitrogen oxides (Fig 7).

Concentration of nitrogen oxides escaping during waste combustion does not exceed specified permissible norms for waste in Lithuania, but in this case CO concentration limits were significantly exceeded. Maximum measured CO value reached 4397 mg/m³. However, CO concentrations are technological parameters, and after changing of combustor, size of pellets or granules or air content permissible CO values can be obtained.

During combustion of selected fuel type (Table 1) no sulfur dioxide escaped, insignificant sulfur dioxide amount was observed in cases when fuel with fuel oil additives.

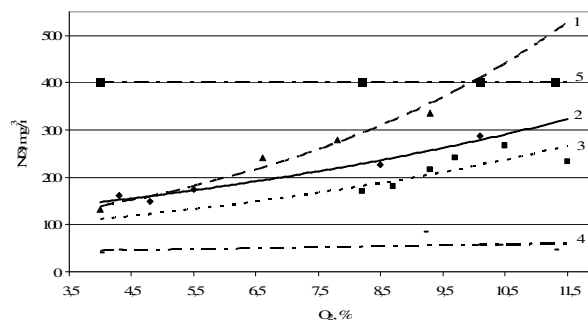


Fig 8. NOx dependence from oxygen during waste combustion: 1 – mixture of all fuel types; 2 – lignin and fuel oil pellets; 3 – straw and fuel oil pellets; 4 – shredded packs (tetrapacks); 5 – concentration rates for NOx

During combustion of straw and fuel oil there is good gasification combustion, high temperature. After combustion ash is observed.

Lignin and fuel oil combustion is very intensive; there is very high flame temperature in combustion (1200 – 1350 °C). Ash is carbonized on grids and cleaning is required. Lignin has very good combustible properties, norms for CO concentration can be achieved in all cases, however, the more fuel oil is used, the more ash is produced. Figure 9 includes measured NOx and CO concentrations during combustion of lignin and fuel oil pellets.

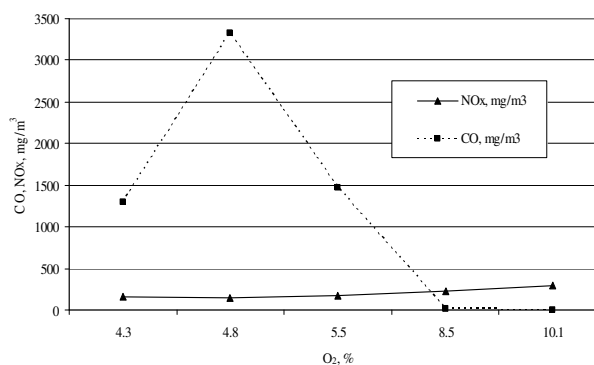


Fig 9. NOx and CO dependence from oxygen during lignin and fuel oil pellets combustion

Huge ash content and complete combustion was observed during combustion of shredded packs (tetrapacks). Fuel is light, although there is a lot of ash on grids, required amount of air is supplied. Fuel is combusted easily, it is proved by contamination concentrations (Fig. 10). Determined optimal oxygen concentration value is about 4,3 %. In this case there are the lowest nitrogen oxide concentrations - about 45 mg/m³, and CO - 246 mg/m³.

Mixture of all fuel types include a lot of waste, i.e. fuel oil, straw, diesel fuel waste, etc. During combustion of such fuel very high temperature was reached. So, there was solid carbonization of grids (Fig 11). It is necessary to remove coke from grids frequently, so it is difficult to feed air. The fuel is combusted quickly and fuel supply speed increases. Flame temperature is high, little ash, so air amount required for combustion cannot be decreased, then unburned fuel amount increases.

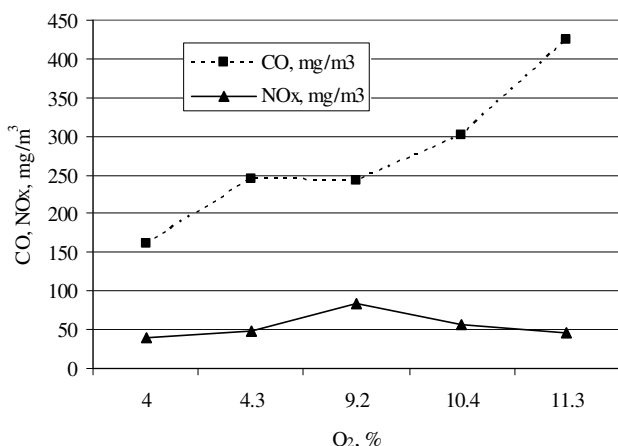


Fig 10. NOx and CO dependence from oxygen during shredded packs (tetrapacks) combustion

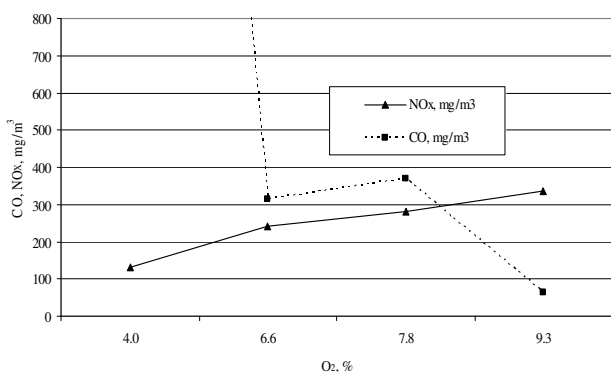


Fig 11. NOx and CO dependence from oxygen during mixture of all fuel types combustion

NOx concentration dependence on oxygen content has well-expressed maximum. During change of oxygen concentration from 5 % to 9,5 % NOx concentration varies from x1,5 to x2 in case of combustion of cornstarch pellets and corn stalk pellets.

Statement that “in case of CO concentration decrease, NOx concentration significantly increases” was proved by experimental method (Wandrasz and Pikon 2007).

Highest oxide concentration in combustible fuel leads to higher NOx concentrations. This statement was proved by investigations (Fig 2 and 8) and it was determined that NOx depends on nitrogen N content in combustible material – the more nitrogen, the more nitrogen oxides in end products of combustion.

To sustain the theory nitrogen content was artificially increased in combustible wood pellets – by adding different carbamide and water solutions. Sprayed pellets were dried naturally and combusted (Fig 12).

Oxide concentrations measured using experimental method conformed to biofuel requirements, except the case when pellets were sprayed artificially by carbamide ($N_K = 2,5\%$). CO values did not reach 2000 mg/m^3 and did not exceed permissible values applied in Lithuania.

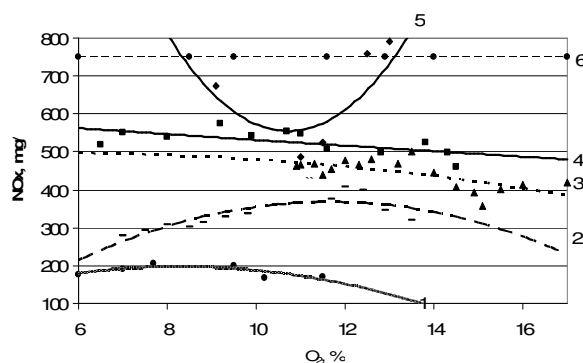


Fig 12. NOx dependence from oxygen: 1 – wood pellets, when $N_K = 0,13\%$; 2, 3, 4, 5 – wood pellets with carbamide ($N_K = 0,3; 0,5; 1,0; 2,5\%$); 6 – concentration rates for NOx

After nitrogen content was increased, nitrogen oxide and carbon monoxide content was also increased, combustion became difficult because of coke formation, and grids had to be cleaned. Flame temperature was very high, little ash; however, air amount required for combustion process cannot be reduced, so the amount of unburned fuel increases.

Figure 13 provides nitrogen conversion dependence on nitrogen content in fuel. Calculations are performed applying formula 3. For the purpose of comparison the diagram includes K_N values obtained during other investigations: points 0,1; 0,4; 0,65 – during combustion of kerosene with pyridine (C_5H_5N) additives (Буйнявичюс and Цирульников 1990); points 0,22; 0,44 – during combustion of natural gas with karbamide additives (Bui-nevičius 2009); point 2,03 – during combustion of dry pig manure (Čepanko *et al.* 2008). Other points are found in Table 1.

It can be seen from the diagram, that there more nitrogen content is, the less nitrogen is converted into nitrogen oxides.

The analysis of investigation results showed that if there are totally different nitrogen sources and different types of combustible fuel, contained nitrogen conversion into NOx can be generalized by the following equation:

$$K_N = 11,312 \cdot N_K^{-0,6196}, \quad (4)$$

here: K_N – conversion factor of nitrogen contained in fuel, percentage; N_K – nitrogen content in fuel, percentage; 11,312 – conversion factor.

Figure 14 provides dependence of nitrogen oxides on oxygen during wood pellets with different nitrogen content combustion.

Provided for under sections in to 7 points; 9; 10,5; 12 (Fig 14), we see, that there more the oxygen content is, the less nitrogen is converted into NOx. For example: when to fuel is added 0,17 % nitrogen to 12 % oxygen contributed 249 mg/m^3 of nitrogen oxides.

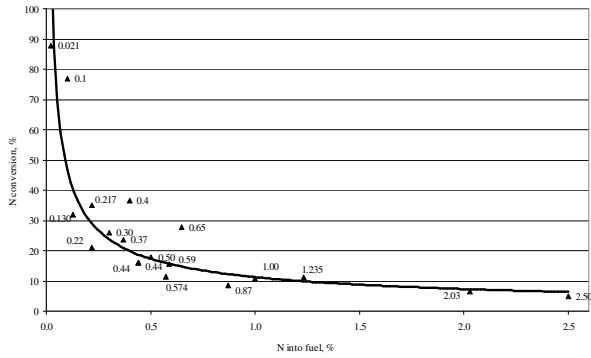


Fig 13. Nitrogen part contained in the fuel which is converted into nitrogen oxides

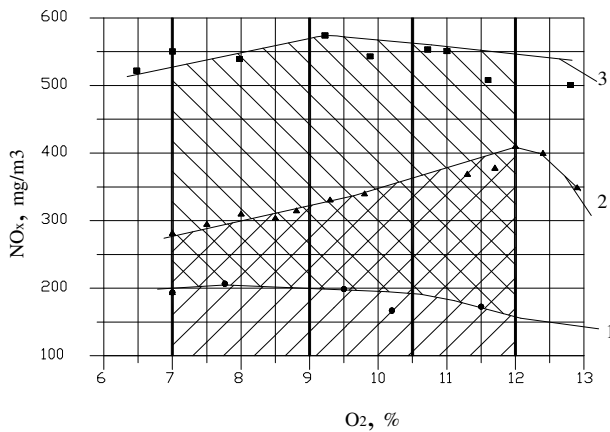


Fig 14. "Fuel" nitrogen oxides dependence from oxygen: 1 - 1 - obtained during wood pellets combustion, when $N_K = 0,13\%$; 2, 3 –wood pellets with carbamide ($N_K = 0,3; 0,5\%$)

At 12 % amount of air NOx was 376 mg/m³. That means, 249 mg/m³ is the real "fuel" nitrogen oxides, and the remaining 127 mg/m³ is "rapid" NOx, because the "thermal" NOx does not exist here.

Figure 15 provides dependence of nitrogen conversion on the amount of air supplied during combustion.

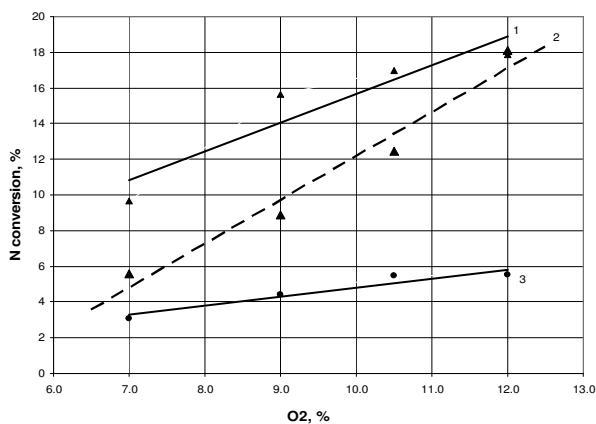


Fig 15. Dependence of nitrogen conversion on oxygen supplied to smoke duct during combustion process: 1 - wood pellets, when $N_K = 0,13\%$; 2, 3 –wood pellets with carbamide ($N_K = 0,3; 0,5\%$)

Although increasing the amount of air, the nitrogen content of the fuel increases twice, but the conversion of nitrogen oxides to nitrogen increases only partially (Fig 15).

4. Conclusions

1. Under the conditions of testing stand CO concentration did not exceed the permissible norms for biofuel. During waste combustion maximum measured value of CO was 4397 mg/m³.
2. During combustion of cornstarch granules NO_x concentrations reached from 482 up to 713 mg/m³, however, the permissible norms for biofuel were not exceeded. Minimum measured value of NO_x concentration during combustion of wood pellets (about 205 mg/m³).
3. During combustion of waste with oil products containing nitrogen the waste requirements were observed. Minimum concentrations were measured during combustion of shredded packs (up to 85 mg/m³), maximum – during combustion of fuel mixture (up to 335 mg/m³).
4. During combustion of wood pellets sprayed with carbamide, nitrogen oxide concentrations did not exceeded the applicable requirements, except fuel combustion, containing $N_K=2,5\%$.
5. Experiments demonstrated that that the more is nitrogen content in the fuel, the less part converts into nitrogen oxides. For example: if nitrogen content in fuel is about 2 %, about 8 % of nitrogen is converted into NOx. However, if nitrogen content in combustible fuel is 0,2 %, about 30 % of nitrogen is converted into NOx. When nitrogen content decreases, i.e. <0,1 %, conversion degree reaches 90 %.
6. The analysis of investigation results showed that if there are totally different nitrogen sources and different types of combustible fuel, contained nitrogen conversion into NOx can be generalized by the following equation: $K_N = 11,312 \cdot N_K^{-0,6196}$.

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