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## REDUCTION OF NITROGEN OXIDES FROM BOILER FLUE GASES

The research carried out on real gases testified to the possibility of intensifying the denitrification of flue gas from coal dust and grate boilers. This was done through the activities aiming at nitrogen oxides removal within the burning zone due to urea injection. The experiments allowed us to establish the influence of powdered urea addition to the coal dust boiler's burning zone on the concentration of nitrogen oxides in flue gases as well as the influence of urea/nitrogen oxide molar ratio on the effectiveness of flue gases denitrification using the mixtures of urea and hydrated lime or calcium carbonate. The influence of urea/nitrogen oxide molar ratio on the quantity of flue gases emitted from grate boiler and the influence of urea solution concentration on the extent of denitrification were also determined.

### 1. INTRODUCTION

As carbon shall long be the main source of energy in Poland [1], it is important to undertake various activities aiming at possibly environmentally friendly usage of carbon fuel in different branches of economy, especially to produce electric energy and heat.

The comparison of different branches of Polish industry in terms of sulphur dioxide and nitrogen oxides emissions in the years 2000 and 2005 shows that the power industry is the main and most severe source of those pollutants (the table). Heat engineering and transport also contribute much to these emissions [1].

The total power of all sources of electric power emission in Poland approaches 32 GW<sub>e</sub>. Heat plants together with commercial heat and power plants produce about 27 GW<sub>e</sub> of that electric power, burning more than 40 million tonnes of hard coal and 65 million tonnes of brown coal. The production of electric energy in Poland comes mainly from 120 to 500 MW<sub>e</sub> condensation blocks with an average efficiency of 33.1%. The majority of energetic blocks are 200 MW<sub>e</sub> blocks, which constitute 40% of the installed power [2].

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Table

## Sulphur dioxide and nitrogen oxides emissions in Poland [1]

Emission source	Sulphur dioxide			Nitrogen oxides		
	2000	2005		2000	2005	
	Gg		%	Gg		%
Commercial power engineering	805	642	52.5	237	247	30.4
Industrial power engineering	265	209	17.1	81	90	11.1
Industrial technologies	91	56	4.6	109	54	6.6
Stationary sources <sup>a)</sup>	309	314	25.7	97	109	13.4
Mobile sources	41	2	0.1	314	312	38.5
<b>Total</b>	<b>1510</b>	<b>1222</b>	<b>100.0</b>	<b>838</b>	<b>811</b>	<b>100.0</b>

<sup>a)</sup> Local boilers, home furnace.

Unsatisfactory technical condition of a great number of blocks, which results mainly from exceeding the planned working time, has led to the application of the following rule to the fuel-energetic sector: specific pro-ecological activities should create a coherent system allowing the optimal ecological effect to be achieved. The influence of power plant or heat and power plant on all the elements of the natural environment should be taken into account.

Nowadays, the necessity of reducing sulphur dioxide and nitrogen oxides emissions to the atmosphere does not raise any doubts. However, the problem of choosing appropriate technologies to clean flue gases is disputable. The reasons for this are different physico-chemical properties of both pollutants and their small quantities in large streams of flue gas. The difficulties in carrying out the cleaning process are additionally affected by the fact that the content of sulphur dioxide in flue gases depends mainly on the type of fuel, whereas the amount of nitrogen oxides formed in combustion processes depends on the temperature of combustion, reagent (nitrogen and oxide) contact time during combustion, especially in the high-temperature zone, the type of furnace devices, etc. [3].

Recently, much attention in professional world literature is paid to the methods of simultaneous sulphur dioxide and nitrogen oxides removal from flue gases. Most often, however, the familiar methods of separate sulphur dioxide and nitrogen oxides removal from flue gases are modified [3].

The methods of reducing sulphur dioxide and nitrogen oxides emissions [4], developed as a result of research, may be divided into two main groups:

- primary methods: reducing the amount of sulphur dioxide and nitrogen oxides produced in the combustion process,
- secondary methods: removing sulphur dioxide and nitrogen oxides from flue gases.

In Poland, simultaneous removal of sulphur dioxide and nitrogen oxides from flue gases may be successfully carried out with the use of selective non-catalytic reduction, which is based on the urea injection to the combustion chamber [5]–[8].

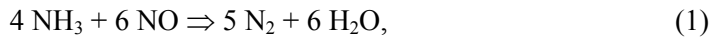
## 2. CURRENT STATE OF TECHNOLOGY

The method of selective non-catalytic reduction of nitrogen oxides from flue gases (*SNCR*) was first patented in 1975 as *Thermal DeNO<sub>x</sub>* [9], then in 1980 as *NO<sub>x</sub>OUT* [10] and in 1988 as *RAPRENO<sub>x</sub>* [11]. Ammonia, urea and cyanuric acid are the reducing agents in these processes. Other reducing agents tested for the nitrogen oxides can be itemized as follows: methylamine, ethylamine, ethylenediamine, and diethylenetriamine [12].

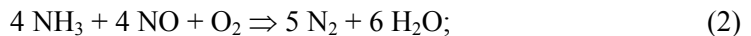
Some of the advantages of *SNCR* are as follows: saving in catalysts and appropriate reactors, saving in flue gas heating (reduction process is carried out in a high-temperature zone) and the possibility of reducing nitrogen oxides emission straight in the source of their production combined with the modification of the boiler and the combustion process.

In the method of selective non-catalytic reduction with the use of ammonia, nitrogen oxides are reduced to nitrogen and water, whereas urea reduces nitrogen oxides to nitrogen, water and carbon dioxide. The overall reaction of the process can be presented as follows:

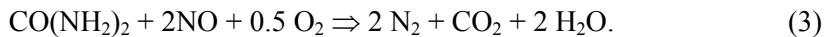
- ammonia



with the use of oxygen



- urea



In reality, the mechanism of nitrogen oxide reduction is far more complex [13], [14].

Cyanuric acid ((HOCN)<sub>3</sub>) decomposes thermally to isocyanic acid (HNCO), which further selectively reduces NO in the exhaust through a multistep chemical reaction mechanism [15].

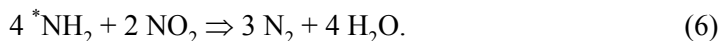
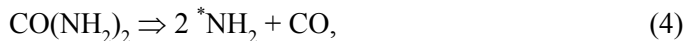
All reducing agents currently used in *SNCR* share a common temperature range, called *the temperature window*, in which the extent of substrate reaction is the highest. A lot of experiments have proved that the temperature window covers the temperature range of 1150–1300 K. Nitrogen oxide concentration in flue gases above these temperatures is high due to ammonia oxidation. At low temperatures, ammonia occurs in excess as a result of incomplete reaction [13]–[24].

In the method of selective non-catalytic reduction of nitrogen oxides with the use of urea, urea solution is injected into the combustion chamber together with the adjunct chemicals, which should reduce temperature to the value necessary for achieving high efficiency of nitrogen oxides reduction. These chemicals are mainly organic sub-

stances disintegrated in the combustion chamber into free radicals:  $^*\text{NH}_2$ ,  $^*\text{OH}$ ,  $^*\text{H}$ ,  $^*\text{O}$  [13], [14].

$^*\text{NH}_2$  radical plays an important role both in the reduction of nitrogen oxides and in the creation of indirect products (reaction (5)).

The reaction with the use of urea can proceed as follows:



According to the overall reaction (3), to transform 2 moles of NO, 1 mole of urea is necessary. The injection of the urea alone causes a required reaction only in a very narrow temperature range, i.e. from 1223 K to 1320 K [18].

The effectiveness of reducing nitrogen oxides from boiler flue gases depends on such basic process parameters as [6]: reaction temperature, contact time in an appropriate temperature range, the type and doses of reducing substance, mole ratio of reducing agent to nitrogen oxides, mixing of reducing agent and flue gases, and chemical composition of flue gases.

The above-mentioned factors variously influence the effectiveness of flue gas denitrification [15]–[27]. Hence, only finding the impact of these parameters on the real effectiveness of flue gas denitrification may settle the question of using this method under Polish conditions in the technologies of flue gases desulphurization in heat and power plants currently in working order or to be realized in future.

### 3. EXPERIMENTAL CONDITIONS

Significant development of dry methods of flue gases desulphurisation through introducing sorbent to the boiler combustion zone (LIMB, LIFAC, COOLSIDE methods) [28], [29] as well as the impressive results of the research on flue gases desulphurisation with the WAWO method conducted since 1989 at the Wrocław University of Technology in an industrial pilot-scale unit of heat and power plant have inspired the works on the intensification of flue gases denitrification process by the activities aiming at nitrogen oxides removal within the combustion chamber [6], [8].

#### 3.1. COAL DUST BOILER

The research on the selective non-catalytic reduction of nitrogen oxides in flue gases was conducted in the WP-120 boiler, equipped with the installation for flue gases desulphurisation with the WAWO method [30].

Figure 1 presents a simplified model of this installation. The basic elements of the installation are reactors, spraying node for liquid dosing and electrofilter [30].

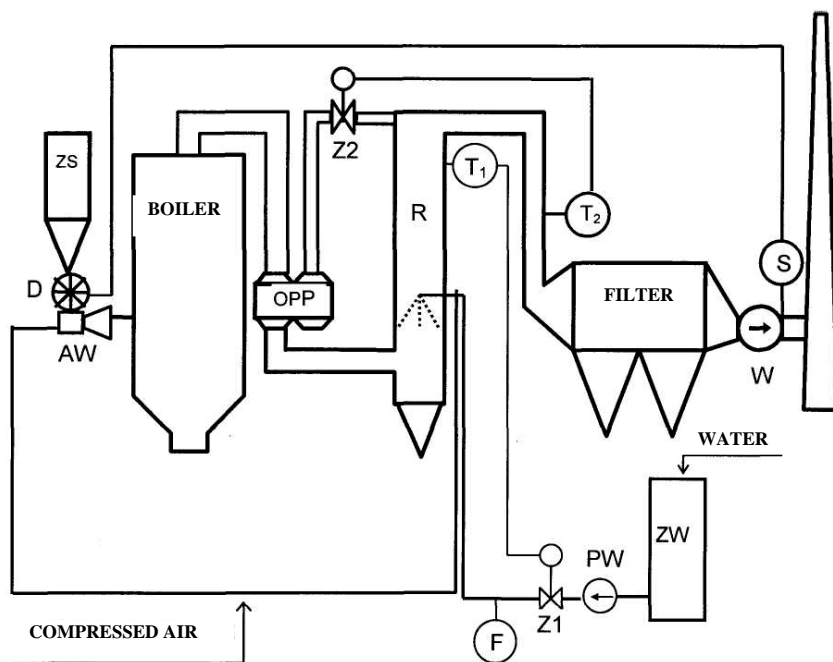


Fig. 1. The model of flue gas desulphurisation and denitrification system:

ZS – calcium (urea) tank, F – flow measurement, R – reactor,

S – SO<sub>2</sub> and NO concentration measurements, ZW – water tank, T – temperature measurement, D – proportioner, PW – water pump, AW – inblowing apparatus, W – fan, OPP – revolving air preheaters

The research carried out with the WAWO method in desulphurisation installations since 1990 revealed that the concentration of sulphur dioxide stabilised on the level of 600 mg/m<sub>n</sub><sup>3</sup> at the following working parameters [30]: flue gas temperature in the reactors: 353 K, flue gas temperature behind the electrofilter: 373 K, the consumption of spraying liquid: 1–16 m<sup>3</sup>/h depending on the power of the boiler, and Ca/S ratio: 2.5/1. The additive application to spraying the reactors with liquid further increased the effectiveness of flue gas desulphurisation. Thus, the use of calcium acetate allowed the outlet concentration of SO<sub>2</sub> in flue gases to be decreased to 450 mg/m<sub>n</sub><sup>3</sup>.

The research on flue gases denitrification was conducted in the WP-120 boiler after its equipment with low-emission burners, i.e. powdered stream burners, which fill the chamber with the flame evenly and reduce the emission of nitrogen oxides in comparison with the rotational burners.

Urea dosing node consists of the storage tank, retention tank, and pneumatic transport installation. The urea ground in the clinker-mill into <20 μm fractions is then

blown above the combustion zone into 1223–1323 K temperature zone. The transport between the storage tank and the retention tank is pneumatic. Urea can be transported from the middle tank by two cell feeders to four inblowing apparatuses.

Cell feeders are automatically steered depending on  $\text{NO}_x$  concentration behind the electrofilter. Switching urea feed into inblowing apparatuses is additionally automatically steered depending on the boiler load. It is connected with the migration of the required temperature areas in the boiler as a result of reduced boiler load. Sulphur dioxide and nitrogen oxide concentrations were achieved in the unit of data processing of the Westinghouse automatic flue gas analyzer.

### 3.2. GRATE BOILER

The research was conducted in the furnace chamber of WR-25 boiler, whose flue gases were fed to the industrial installation of flue gas desulphurization [31].

In order to estimate the effectiveness of reducing nitrogen oxide emission from the boiler, the measurement of nitrogen oxide concentration in flue gas behind the boiler in two subsequent situations was carried out: carbon combustion without dosing urea solution and carbon combustion with dosing urea.

The concentrations of sulphur dioxide, nitrogen oxide and oxygen in flue gas were measured with the GA-60 MD MADUR analyzer. Each measuring cycle lasted 60 minutes, as is required by the Polish law for one-hour emission of pollutants [32]. All the measurements were averaged for the period of 60 minutes. Before the essential research, a number of measurements were taken in order to establish the initial parameters of the boiler.

WR-25 boiler is adjusted to the combustion of fine hard coal on a horizontal travelling belt grate. Fuel is gravity-fed from the hopper onto the grate. The stream of the combusted fuel is controlled by the height of the fuel layer and the speed of the grate headway. Fuel gets from the hopper straight beneath the fire vault, where it is moved on the grate and successively dried, degassed and its volatile parts are lit. In the further part of the grate, degassed carbon is combusted. The air for combustion is fed under the grate (primary air). The air is also added above the grate (secondary air) to combust the products of partial combustion: carbon oxide, hydrocarbons and soot.

The dosing node of urea solution consists of the urea solution storage tank, stirrer and the installation for the transport of technological water and compressed air. A saturated urea solution is fed with the pump from the storage tank to the stirrer, where it is diluted to the desired concentration (10 or 40%). Then, it is directed through a flowmeter to injection nozzles spaced on the same level on both sides of the boiler.

In the experiments, urea solution was injected into the combustion chamber above the combustion zone in the temperature area of 1223–1323 K with 2 nozzles, in the

amount of 5 to 30 kg/hr because, during the research, the average nitrogen oxide concentration was  $180 \text{ mg/m}_n^3$  and the average flue gas stream equalled  $55550 \text{ m}_n^3/\text{hr}$ .

## 4. RESULTS AND DISCUSSION

### 4.1. COAL DUST BOILER

The research allowed us to estimate the influence of powdered urea addition to the combustion zone in the boiler with a temperature of 1273 K on the concentration of nitrogen oxide. Figure 2 illustrates the results obtained.

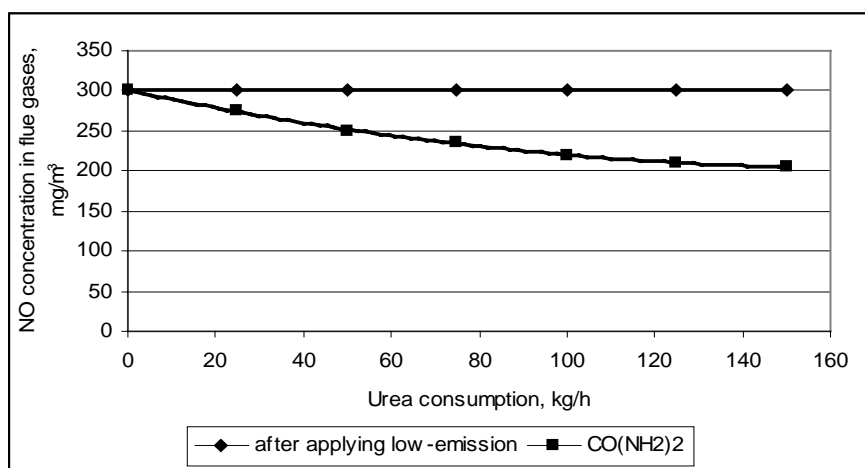


Fig. 2. NO concentration in flue gases versus urea consumption

Figure 2 shows that together with urea consumption during the process, nitrogen oxide concentration in flue gas decreased from  $300 \text{ mg/m}_n^3$  after applying low-emission burners to  $205 \text{ mg/m}_n^3$  with the use of selective non-catalytic reduction.

Additionally, during the experiment, a decrease in the sulphur dioxide concentration by 18–45% was observed, as well as an increase in carbon oxide content. The presence of cyanides in flue gas was not attested. The results of the research obtained so far allow us to state that urea consumption depends greatly on sulphur dioxide concentration in flue gas. Hence, it seems that a more economic way of dosing powdered urea would be feeding it together with calcium compounds ( $\text{CaCO}_3$ ,  $\text{Ca(OH)}_2$ ,  $\text{CaO}$ ), which are used for reducing sulphur dioxide in flue gases emitted from heat and power plants.

The influence of urea/nitrogen oxide molar ratio on the extent of flue gas denitrification with the use of the mixture of urea with caustic lime or with calcium carbonate is presented in figure 3.

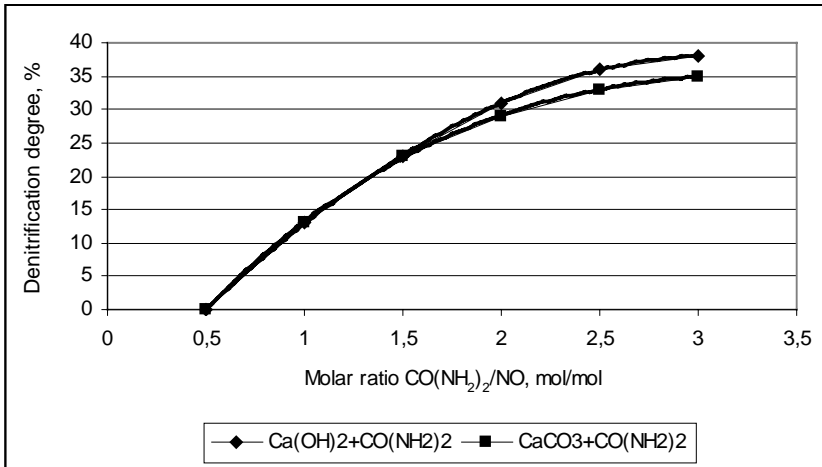


Fig. 3. Denitrification rate versus  $\text{CO}(\text{NH}_2)_2/\text{NO}$  molar ratio for different sorbents:  $\text{Ca}(\text{OH})_2 + \text{CO}(\text{NH}_2)_2$  and  $\text{CaCO}_3 + \text{CO}(\text{NH}_2)_2$

Figure 3 shows that at the ratio of  $\text{CO}(\text{NH}_2)_2/\text{NO} = 1.0$ , 23% of nitrogen oxide was reduced whereas at this ratio being 2.5-fold higher, the degree of a denitrification increased additionally by 15% for the mixture of urea with caustic lime and by 13% for the mixture of urea with calcium carbonate.

The results of using urea in the combustion zone in the boiler as well as the observations of the WAWO installation working with the use of selective non-catalyst reduction prove the appropriate efficiency of SNCR method. They also prove that the method ensures the level of 180–210  $\text{mg}/\text{m}^3$  of nitrogen oxides concentration in purified gases regardless of the boiler load. The method can then be successfully applied to the process of simultaneous flue gas desulphurization and denitrification in heat and power plants.

#### 4.2. GRATE BOILER

Based on the experiments it is possible to estimate the influence of different concentrations of urea solution fed through 2 injection nozzles to the combustion zone at the temperature of 1273 K on nitrogen oxide concentration (figure 4).

The addition of urea during the process decreased nitrogen oxide concentration in flue gas to various values, depending on the concentration of the solution used (fig-



ure 4). Thus, nitrogen oxide concentration in flue gas dropped from  $180 \text{ mg/m}_n^3$  (without urea injection) to  $51.8 \text{ mg/m}_n^3$  for 40% urea solution, and to  $68.8 \text{ mg/m}_n^3$  for 10% urea solution.

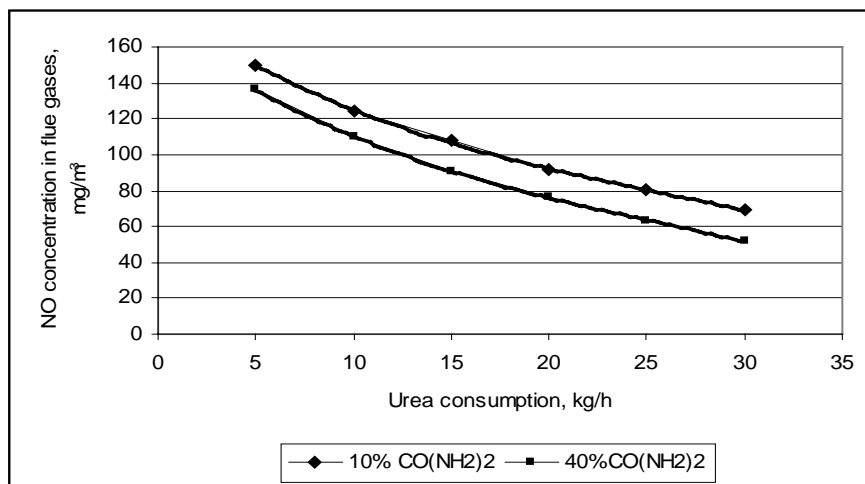
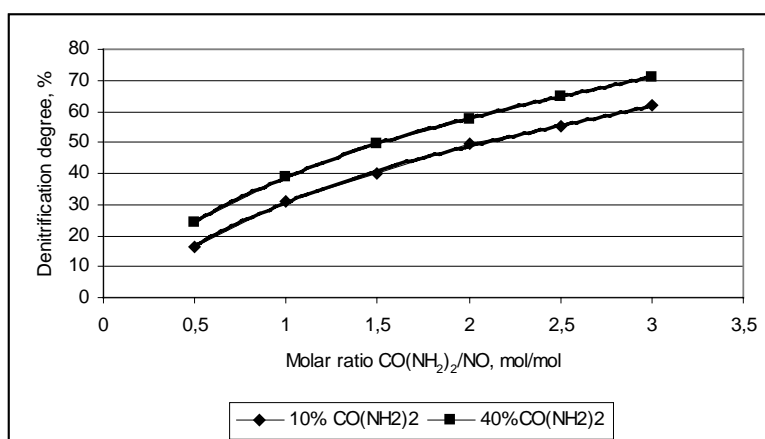


Fig. 4. NO concentration in flue gases versus urea consumption for different concentrations of solutions (10%; 40%)

The influence of urea/ nitrogen oxide molar ratio on the extent of flue gas denitrification, depending on the applied concentration of the solution, is shown in figure 5.



Rys. 5. Denitrification rate versus CO(NH<sub>2</sub>)<sub>2</sub>/NO molar ratio for different concentrations of urea solutions (10%; 40%)

Using 40% urea solution at the ratio of  $\text{CO}(\text{NH}_2)_2/\text{NO} = 1.0$  caused 38.9% nitrogen oxide reduction, whereas at this ratio being 2.5 times higher, denitrification degree increased additionally by 26.1% (figure 5).

During the experiments, 15–34% reduction of sulphur dioxide in flue gas was additionally observed as well as 15–22  $\text{mg}/\text{m}^3$  increase of carbon oxide in flue gas (figure 6).



Fig. 6. CO concentration in cleaned flue gases versus urea consumption

A selective non-catalyst reduction of nitrogen oxides from flue gas in grate boiler proved explicitly that the higher the amounts of urea, the more effective the flue gas denitrification. The maximum 71.2% effectiveness of flue gas denitrification was achieved at the injection of 40% urea solution and the molar ratio of  $\text{CO}(\text{NH}_2)_2/\text{NO} = 3.0$ .

Previous experiments allow us to conclude that the degree of flue gas denitrification may be further increased by optimizing the injection of urea solution through the use of a maximum number of injection nozzles and their changeable setting angle so that the urea spraying covers, if possible, the whole horizontal surface of the furnace chamber.

## 5. SUMMARY OF RESULTS

The results of the research using urea in the burning zone in coal dust and grate boilers indicate that this method can successfully be applied to the process of simultaneous flue gas desulphurisation and denitrification in heat and power plants and boiler houses.

Literature data [16]–[19], [23]–[27] prove that the increase of nitrogen oxide reduction with urea causes the rise in an excessive ammonia concentration in flue gas at

the boiler outlet. Under ordinary operating conditions, the ammonia concentration in outlet flue gas approaches 1–5 mg/m<sup>3</sup>.

In the research conducted, the concentration of an excessive ammonia was not analyzed because WAWO installation [30] has a built-in reactor behind the coal dust boiler, in which flue gases are sprinkled with alkaline solution, and DAM installation [31] fixed behind the grate boiler is able to reduce an excessive ammonia (10–25 mg/m<sup>3</sup>) to acceptable values.

The increase of carbon oxide concentration in flue gas observed during the experiments is an undesirable side-effect. One of the factors determining the amount of the urea solution dosed should be the concentration of carbon oxide in flue gas sustaining lower than its acceptable concentrations [32].

The results of the research indicate that the use of selective non-catalyst reduction method to decrease nitrogen oxides in flue gas emission with the use of urea solution has proved to be fully successful. This method allows us to operate coal dust and grate boilers at relatively low investment costs [33] maintaining the emission of toxic substances from flue gas according to the emission standards [32].

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