# ARCHIVESOFENVIRONMENTALPROTECTIONA R C H I W U MO C H R O N YŚ R O D O W I S K Avol. 30no. 1pp. 21 - 322004

PL ISSN 0324-8461

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# THE INFLUENCE OF THE TYPE AND QUALITY OF COAL ON $\mathrm{NO}_{\mathrm{X}}$ EMISSIONS DURING PULVERISED COAL COMBUSTION

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Keywords: nitrogen oxide emission, power engineering, coal combustion.

# WPŁYW TYPU I JAKOŚCI WĘGLA NA EMISJĘ NO<sub>x</sub> W CZASIE SPALANIA PYŁU WĘGLOWEGO

W laboratoryjnym piecu z quasi-jednowymiarowym przepływem laminarnym przeprowadzono badania wpływu własności węgla na emisję tlenków azotu dla 23 węgli energetycznych w zakresie stopnia uwęglenia od węgli brunatnych poprzez węgle kamienne do antracytów. Dla jednego typu węgla brunatnego dwukrotny wzrost zawartości azotu powodował wzrost emisji NO<sub>x</sub> o 30%. Ze wzrostem stopnia uwęglenia, od węgli brunatnych do kamiennych, następował wzrost emisji NO<sub>x</sub> i następnie jej spadek dla antracytów. Wzrost zawartości azotu powodował wzrost emisji tlenków azotu i spadek stopnia konwersji azotu zawartego w paliwie. Na podstawie wyników badań i ich analizy statystycznej określono zależności opisujące wielkość emisji NO<sub>x</sub> i stopień konwersji azotu zawartego w paliwie do tlenków azotu w funkeji dwóch zmiennych, opisujących własności paliwa, tj. zawartości azotu i wskaźnika paliwowego definiowanego jako iloraz zawartości stałych części palnych i zawartości części lotnych.

#### Summary

In laboratory electrically heated entrained flow reactor with quasi one-dimensional laminar flow the influence of fuel properties on the nitrogen oxide emission for 23 coals of various rank from brown coal, bituminous coal to anthracites has been examined. For one brown coal type twofold increase of nitrogen content resulted in NO<sub>x</sub> emissions increase of about 30%. Increasing rank from brown coal to bituminous coal increases NO<sub>x</sub> emissions and next a fall in the anthracites range. With nitrogen content increase NO<sub>x</sub>-emission grows and the ratio of fuel nitrogen conversion to nitrogen oxide decreases. Experiments results and their statistical analysis were used to determine relationships describing NO<sub>x</sub> emissions and conversion ratio of fuel nitrogen to nitrogen oxide as functions of two variables, describing coal properties, i.e. nitrogen content and fuel ratio (fixed earbon to volatile matter ratio).

#### INTRODUCTION

The basic source of NO<sub>x</sub> emissions – constituting 75–95% of total NO emission from pulverised coal flame – is the oxidation of nitrogen chemically bonded with fuel. The dominating role of fuel NO<sub>x</sub> in coal combustion comes from the fact that combustion temperatures are relatively moderate (1500–2000 K) and local zones are rich in fuel mainly pulverised coal fuel. Due to complex reactions between aerodynamics near the burner, fuel

oxidation and chemical reactions mechanism aimed to form nitrogen compounds, there are no complete rules defining the mechanism of  $NO_x$  formation for a given configuration of a burner and a combustion chamber.

Thus it follows that the type and quality of burned coal, as well as operational conditions [10], play a major role in NO<sub>x</sub> formation. NO<sub>x</sub> formation depends on such coal qualities as nitrogen and volatile matter contents, rank and the division of fuel nitrogen compounds into volatile matter and char residue. It is estimated that nitrogen compounds contained in volatile matter form 60 to 80% of nitrogen oxides [13, 15]. Although intensive research has been conducted on the mechanism of NO<sub>x</sub> formation from pulverised coal furnaces, sufficient data characterising NO<sub>y</sub> formation rate and its relation to type and quality of coal do not exist yet.

The subject of this work is establishing basic rules and general characteristics related to NO<sub>x</sub> formation from the combustion of various coals in the form of pulverised coal in technical flame by describing the influence of fuel properties (type and quality of coal). On the basis of statistical analysis an attempt will be made to determine empirical relation, according to which it will be possible to predict NO<sub>x</sub> emissions from the combustion of various coals in nonstaged combustion. Basic information obtained in this study may be applied to optimise real systems with low NO<sub>x</sub> emission by implementing appropriate local combustion conditions. Particular goals involve:

- Identification and quantity description of coal properties which influence NO<sub>x</sub> emissions,
- Ranking coals in the order of their potential to form NO.

# COAL TYPE INFLUENCE ON $\mathrm{NO}_{\mathrm{x}}$ EMISSIONS

The selection of coal for a power plant usually involves costs analysis or the reduction of  $SO_2$  emission. The change of coal will also influence  $NO_x$  emissions level and the loss of efficiency due to incomplete combustion. Coal properties have impact on NO reduction in operational conditions with air staged because it is mainly nitrogen released with volatile matter to be sensitive to substoichiometric conditions in flame. In combustion with nonstaged air condition, the transformation of nitrogen (released with volatile matter) to NO is more effective than the conversion of nitrogen which remained in char residue. That is why the increase of volatile matter content in fuel (combustion of low rank coals) results in higher NO emission in nonstaged combustion [11, 17]. Usually differences in coal rank cause differences in temperature and residence time in fuel-rich zone, in consequence this indirectly influences the quantity of nitrogen compounds and reaction rate.

The connection between nitrogen content in fuel and NO emission is very misleading. A general observation has been made that the higher nitrogen content in coal, the higher NO emission was registered [3, 6]. However, nitrogen content in coal is not used as an indicator of potential NO emission from a given fuel. Morgan [11] observed a distinct correlation between nitrogen content in volatile matter and NO emission in nonstaged air combustion. In operational conditions with staged air there was no such relation and differences between NO emission value from particular coals were minor. Chen *et al.* [3] presented results for 48 coals in the form of relation between NO emission grew with the increase of nitrogen content in coal. In the combustion of low rank coal, usually containing less nitrogen, NO emission was lower. However, within the given rank fuel properties

other than fuel nitrogen content also mattered. If we take the example of brown coal containing about 1.05% of nitrogen, NO<sub>x</sub> emissions values differed by 60%.

Generally, it was observed that low NO<sub>x</sub> emissions take place in rapid coal devolatilisation conditions in fuel-rich zone; in a situation when nitrogen compounds are released with volatile matter they will transform to N<sub>2</sub> instead of NO<sub>x</sub>. That is why coals with high content of volatile matter have bigger potential for low NO<sub>x</sub> emissions. Anthracites show lower emission in nonstaged combustion because the prevailing part of fuel nitrogen remains in char residue, besides fuel nitrogen content is lower in anthracites than bituminous coal. In staged air combustion, final NO<sub>x</sub> emission reduction will be less effective for coals, for which a significant nitrogen part remains bonded with char residue.

The results of research on the influence of coal type which were conducted on a large scale have been presented in the works of Benesch and Schadt [1], Koda *et al.* [8], and Tigges *et al.* [18]. Tigges *et al.* [18] examined the influence of volatile matter content on NO<sub>x</sub> emission from a boiler. NO<sub>x</sub> emission decreases with the increase or the reduction of volatile matter content from the level of 25%. Koda *et al.* [8] conducted research on a power unit of 1000 MWe with wall burners for 8 different coals varying fuel ratio, FR (FR is defined as the ratio of fixed carbon to volatile matter content), nitrogen and ash contents, as well as particle size. On the basis of the research the following conclusion was made: low fuel ratio values generated lower NO<sub>x</sub> emissions and solid residue. It was observed that the increase of nitrogen content in fuel increased NO<sub>x</sub> emission only to a lesser extent. Benesch and Schadt [1] explained this notion by the fact that coals with high nitrogen content show lower transformation rate of fuel nitrogen to fuel NO<sub>x</sub>, this is due to slow release of nitrogen compounds in comparison with coals containing less nitrogen.

Some coal types are better for combustion with the application of various techniques reducing  $NO_x$  emission. Due to the danger of corrosion, coals with high sulphur content are not applicable for combustion in substoichiometric conditions used in staged combustion. Grusha and Hurt [5] conducted research in a boiler with tangentially burners where they burned 4 different coals. They made an observation that the amount of formed  $NO_x$  increased with the growth of stoichiometric ratio. However, coals with high volatile matter contents and low FR value generated lower  $NO_x$  emissions for the same values of stoichiometric ratio regardless of nitrogen content in coal. It can be concluded, that in order to obtain maximum  $NO_x$  reduction, combustion stoichiometry optimisation it may be necessary to match combusted coals.

## EXPERIMENTAL AND COAL SAMPLES DESCRIPTION

#### Coal samples and their physic-chemical analyses

Proximate and elemental analyses in connection with calorific value determination constitutes a basic test in the classification of coal rank for a given coal sample. The research was conducted on 23 coal samples varied in rank, from brown coal, bituminous coal to anthracites. The influence of coal quality changes was examined for 6 coals, varying in ash contents and calorific value, originating from one type of brown coal. Coal samples were analysed in laboratory conditions regarding the following factors: moisture content, ash, volatile matter, fixed carbon (FC), calorific value (high-heat value,  $Q_s$ ), the content of the following elements (C, H, S, N). Research results are presented in Tables: 1 (proximate analysis) and 2 (ultimate analysis).

No	Coal sample designation	Moisture	Ash	Volatile Matter	Fixed Carbon	High-heat value	Low-heat value					
		Wa	Aª	Vª	FCª	$Q_{sp}^{\ a}$	$Q_i^{\ a}$					
		%	%	%	%	kJ/kg	kJ/kg					
Brown Coals												
1	AD	15.7	18.2	41.8	24.3	17835	16487					
2	В	4.4	22.2	42.1	31.3	20871	19803					
3	KO	11.4	39.5	30.4	18.8	13582	12308					
4	WL	13.8	19.6	43.1	23.4	18594	17032					
5	T1	8.7	34.3	33.5	23.6	15622	14537					
6	T2	11.1	24.2	37.0	27.6	18773	17432					
7	T3	10.6	39.6	30.1	19.8	12732	11643					
8	T4	16.5	9.5	40.8	33.1	21987	20295					
9	T5	10.2	49.9	25.3	14.6	10008	9060					
10	T6	10.5	28.3	36.1	25.1	17062	15780					
Bituminous Coals and Anthracite												
11	JW	8.5	33.6	24.8	33.1	17528	16549					
12	KT	3.9	11.6	28.4	56.1	29116	28003					
13	NI	4.6	27.5	23.8	44.1	20500	19720					
14	OD	1.5	21.9	27.8	48.8	25936	25016					
15	OP	2.6	13.2	30.3	53.9	28135	27110					
16	PO	3.8	22.5	28.1	45.6	23716	22867					
17	SK	1.8	20.9	26.1	51.2	26310	25465					
18	SR	7.7	26.1	27.2	39.0	19796	18768					
19	WR	3.1	26.7	24.1	46.2	23666	22661					
20	MD	3.6	18.5	20.2	57.7	26384	25346					
21	ELC	2.1	9.6	36.9	51.4	32366	31110					
22	IBB	4.6	17.9	6.4	71.1	30281	29528					
23	ANT	1.7	3.1	4.9	90.3	34032	33219					

Table 1. Proximate analysis results (analytical state)

#### Experimental study and research procedure

A problem which is very often the case in the research aimed at analysing the influence of various parameters is the fact that a change in one parameter leads to undesired changes in another parameter. That is why the research on the combustion of the above listed coals was carried out in laboratory conditions in an electrically heated reactor with controlled temperature and with quasi one-dimensional flow. Such conditions make it possible to perform an independent determination and examination of coal type and quality, stoichiometric ratio, temperature and particle size. The following range of operational conditions changes was possible to be obtained in the equipment (the studies of the influence of the type and coal quality on NO, emission are conducted for conditions given in breaks):

- stoichiometric ratio value from 0.8 to 1.8, (1.0–1.4);
- reactor temperature values: from 1073 to 1273 K, (1173 K);
- particle size below 200 μm, coal was ground to size such that at least 70% passes a 80 μm screen.

No	Coal sample designation	Analytical State (wt%)								
		Cª	Hª	Nª	Sª	Oª				
Brown Coals										
1	AD	45.5	4.4	0.70	1.58	13.9				
2	В	46.0	4.4	0.54	1.00	21.5				
3	KO	34.4	4.6	0.44	0.44	9.3				
4	WL	38.7	5.6	0.48	0.66	21.1				
5	T1	37.6	4.0	0.57	1.26	13.6				
6	T2	45.1	4.9	0.54	0.68	13.4				
7	T3	32.1	3.8	0.46	0.84	12.6				
8	T4	54.9	5.9	0.70	0.55	11.9				
9	T5	24.6	3.2	0.38	0.72	11.0				
10	T6	41.7	4.7	0.56	1.32	13.0				
Bituminous Coals and Anthracite										
11	JW	42.6	3.5	0.82	1.34	9.6				
12	KT	69.5	4.7	1.32	0.62	8.4				
13	NI	50.7	3.1	0.99	1.40	11.7				
14	OD	60.3	4.0	1.10	1.00	10.1				
15	OP	68.1	4.4	1.10	0.65	9.9				
16	PO	51.2	3.5	0.71	1.20	17.2				
17	SK	59.7	3.7	0.92	1.30	11.8				
18	SR	46.2	3.8	0.97	1.50	13.7				
19	WR	55.8	4.3	0.99	1.12	8.0				
20	MD	65.3	4.4	1.66	0.44	6.2				
21	ELC	70.4	5.5	1.41	0.56	10.3				
22	IBB	89.0	3.5	1.22	0.45	1.0				
23	ANT	70.4	2.9	1.01	1.00	2.2				

Table 2. Ultimate analysis results

Research on  $NO_x$  emission was conducted [10] during nonstaged combustion of pulverised coal on a test stand consisting of a vertical reaction chamber, a flat kinetic pulverised coal burner on the inlet of the chamber, pulverised coal feed system with a fluidised coal feeder, electric heating system with temperature control for a combustion chamber, cooling system, flue gas and air systems, measurement system for flue gas composition and current stoichiometric value.

The reaction chamber measured 900 mm in height, it was made from a quartz tube 55/47.8 mm in diameter. Electrically heating system with temperature control ensured the required level of temperature. A flat burner can operate in a stable manner thanks to a kinetic flame of propanebutane and air mixture. Pulverised coal is fed with air through a fluidised coal feeder. The feeder is a system of nozzles which feed carrying air, outlet system and pulverised coal container. A fluidised coal feeder is placed on laboratory scales. The information of current pulverised coal and feeder weight is read every 2 s by a computer connected to the scales, this forms the basis to determine the current pulverised coal flux. Pulverised coal flux is controlled by overpressure regulation system in the feeder and carrier air flow. Flue gas is analysed continuously by a flue gas analyser, they are also registered when the whole system reaches the steady state. The analyser takes the measurement of the content of oxygen, carbon monoxide, carbon dioxide, sulphur dioxide and nitrogen oxides in flue gas. Stoichiometric ratio was determined on the basis of coal mass fluxes, air and propane-butane fluxes, as well as fuel properties. Unburned solid residue was corrected in the calculation of stoichiometric ratio by direct measurement of oxygen and carbon dioxide concentrations in flue gases.

Ranking coals in the order of its potential to form  $NO_x$  was based on statistical analysis, namely regression analysis of the measured values for  $NO_x$  emission with the properties of the examined coals. Regression analysis is an effective tool used to describe dependencies between measured values of various parameters especially in the cases when their individual influence cannot be distinguished quantitatively.

#### RESEARCH RESULTS

## The influence of coal quality on NO<sub>x</sub> emissions

A quality variation of a given coal type is understood as a change in coal properties within the range of one mine. Substantial variations in coal quality can be observed in the case of brown coal properties analysis due to the fact that since brown coal is an intermediate stage in the coalification process, so its properties change more significantly, e.g. moisture and ash content, volatile matter, elemental constitution of combustible and mineral substances. An analysis of coal quality influence on NO<sub>x</sub> emission was conducted on the basis of the examination of 6 brown coal samples from Turów mine. These are samples marked T1 through T6 in Tables 1 and 2.

The examined brown coals differ in: ash content (9.5–49.9%), moisture content (8.6– 16%), volatile matter (25–40%), coal element (24–54%), hydrogen (3.2–5.9%), nitrogen (0.38–0.7%) and low-heat value (9.0–20.2 MJ/kg) – all data were calculated to analytical state. Fig. 1. presents variations in nitrogen oxides emissions as a function of nitrogen content in coal for three different values of stoichiometric ratio. Emission growth was observed with the increase of nitrogen content in fuel although emission variations are considerably smaller in comparison with variations in nitrogen content. Nearly twofold increase of nitrogen content in coal caused emission increase by some 30%. This confirms the fact that other coal properties influence nitrogen oxides emission in combustion.

#### Coal type influence on NO<sub>x</sub> emissions

The research was conducted on coal type influence on NO<sub>x</sub> emissions with coals varying in rank from the lowest rank (brown coal) over bituminous coal to anthracite. The measured for stoichiometric ratio equal 1.1. NO<sub>x</sub> concentrations values were examined in relation to basic coal properties: moisture content, ash content, nitrogen content (Fig. 2), volatile matter content (Fig. 3), carbon content and high-heat calorific value. Similar relations were obtained for the fraction of fuel nitrogen converted to NO<sub>x</sub>, denoted as conversion ratio (CR) [9]. Conversion ratio is defined as the ratio of NO concentration at the exit of the furnace to that of 100% conversion from fuel nitrogen. Some experimental relationships for CR are presented in Figures 4 and 5.

An analysis of the obtained results shows that nitrogen oxides emissions depend on coal properties. With the increase of moisture and ash content a decrease of nitrogen oxides emissions was observed. The influence, however, is insignificant. The growth of volatile matter content in the range of about 25% of coal mass causes emission increase and when this value is exceeded a decrease of NO<sub>x</sub> emissions is observed (Fig. 3).

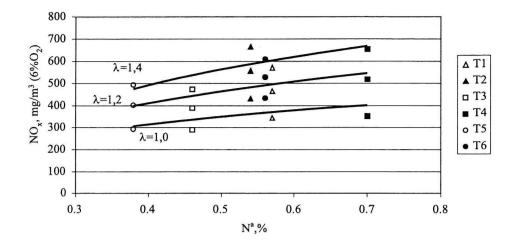


Fig. 1. Variations in nitrogen oxides emissions as a function of nitrogen content in coal for three different values of stoichiometric ratio for one type of brown coal from Turów mine. Furnace temperature 1173 K

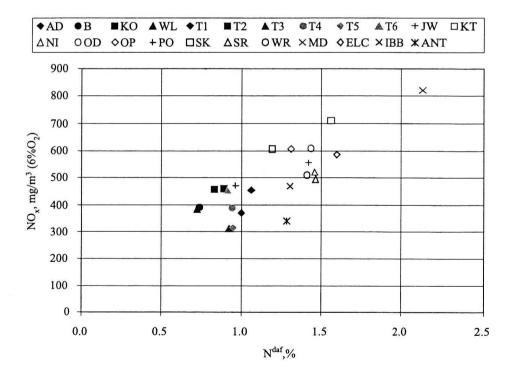


Fig. 2. NO<sub>x</sub> emission index in respect to nitrogen content (daf) for examined coals. Experimental conditions: temperature - 1173 K, stoichiometric ratio - 1.1

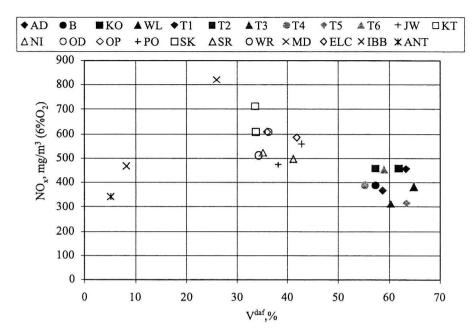


Fig. 3.  $NO_x$  emission index in respect to volatile matter content (daf) for examined coals. Conditions as in Fig. 2

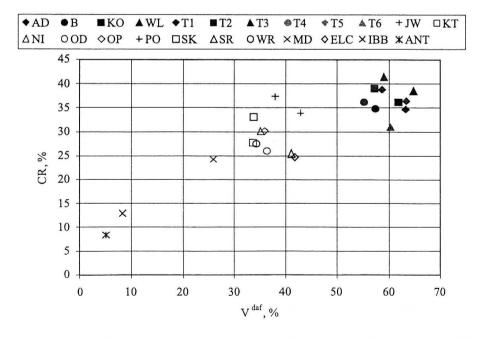


Fig. 4. Conversion ratio of fuel nitrogen to nitrogen oxide in function of volatile matter content (daf) for examined coals. Conditions as in Fig. 2

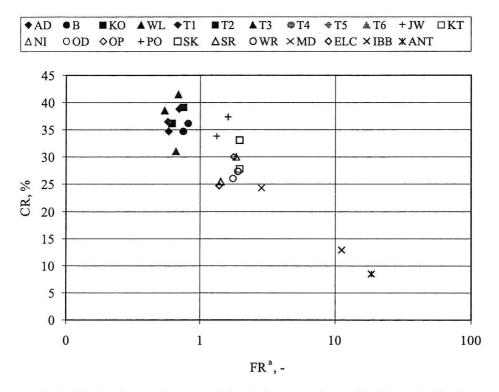


Fig. 5. Relation of conversion ratio to fuel ratio for examined coals. Conditions as in Fig. 2

With the increase in the rank a growth in nitrogen oxides emissions is observed and in the range of anthracite coal and anthracites a decrease was noted. NO<sub>x</sub> emissions relationship to high-heat value has similar progress. Fig. 2 shows that with the increase of nitrogen content in coal NO<sub>x</sub> emission grows. Brown coals (solid symbols are marked in figures) contain less nitrogen and show lower emission than bituminous coals. In some cases in the combustion of coals of similar nitrogen content one can obtain NO<sub>x</sub> emissions varying in over 30%, or even up to 60% [2].

The conversion of nitrogen in char to nitrogen oxides is lower by some 20% in comparison with nitrogen released with volatile matter, in particular conditions with high NO participation even up to about 90% [2]. Older coals contain more carbon and less volatile matter. Anthracites contain minute amounts of volatile matter and over 90% of carbon (daf). That is why emission results for anthracites diverge from the trend and in some cases they must be treated as a separate category.

Figures 4 and 5 show variations in conversion ratio for nitrogen contained in fuel to nitrogen oxides depending on coal properties. Conversion ratio grows with the increase of volatile matter content (Fig. 4.) and decreases with the growth of nitrogen content in coal. The growth in rank and fuel ratio results in the decrease of nitrogen conversion ratio (Fig. 5).

Volatile matter contains such nitrogen compounds as NO, HCN and  $NH_3$  (referred to by the abbreviation TFN – total fixed nitrogen) and insignificant amount of  $N_2$  and sometimes  $N_2O$ . TFN compounds transform to nitrogen oxides in the presence of oxygen [4, 12]. Brown

coals contain the largest amounts of volatile matter, in devolatilization they release most of nitrogen compounds which are transformed to nitrogen oxides to higher extent than those remaining in char. Conversion ratio for older coals is lower just because of the lower content of volatile matter, conversion lowest value is reached in the case of anthracite. Conversion ratio decreases with the growth of nitrogen content in coal [4, 12], which is also connected with the increase of rank decrease of volatile matter content.

Statistical determination of the relation between coal properties and NO<sub>x</sub> emissions

Current literature does not provide us with general relationships which would allow to predict  $NO_x$  emissions in coal boilers. In the past many authors made attempts to relate coal properties, defined on the basis of proximate and elemental analyses, to  $NO_x$  emissions [9, 12, 14, 20]. Recent publications [3, 7] present attempts of prediction  $NO_x$  emissions in the combustion of various coals on the basis of parameters other than proximate and elemental analyses. It has been observed that such parameters as nitrogen content in char residue and the number of nitrogen compounds released with volatile matter may also be taken into consideration as indicators making it possible to predict  $NO_x$  emission from a given coal.

Determining the empirical dependence to be used for NO<sub>x</sub> emission prediction in the combustion of different coals in operational conditions of nonstaged combustion was based on statistical analysis. According to the method applied, first assumptions were made in reference to the number of parameters and descriptive function form, the following stage was determining constant coefficients included in the model. These coefficients may be determined by one of estimation theory methods. The least squares method was applied in the study [19]. Calculation were done by individual software written in FORTRAN with the use of standard procedures [16].

Calculation results show that together the correlation coefficient grows with the increase of the number of parameters. The analysis of parameters significance showed that for the examined dependent variables in respect to predicting their value one should use the following formulas obtained from linear regression:

 $NO_x = 353.25*N^a - 14.95*FR^a + 226.51 \text{ [mg/m^3]},$  (1)

 $CR = -1.471 * FR^{daf} - 10.36 * N^{daf} + 46.84 \quad [\%], \tag{2}$ 

where N is fuel nitrogen content, FR is fuel ratio (defined as ratio of contents of fixed carbon to volatile matter, it means that FR does not depend on bases). The a bove formulas were determined for stoichiometric ratio equal 1.1. Error analysis shows that an average determination error amounts to 9–10% for both variables. Maximum error for conversion ratio amounts to 24% (ANT coal – anthracite) and 30% (IBB coal – anthracite coal). Other errors do not exceed 21% (for both emission and conversion ratio). The formulas shows, that NO emission and conversion ratio are related to nitrogen content and coal rank, where both factors are mutually independent, and that for low rank coal (low FR), nitrogen content was the most significant factor describing emissivity.

#### CONCLUSIONS

The aim of the present study was determining basic properties of  $NO_x$  formation and the influence of coal properties on the process. To achieve the purpose 23 different coals

were burned in laboratory conditions, in electrically heated reactor with controlled atmosphere. As a result of the research individual influence of combusted coal properties on NO formation and fuel nitrogen conversion ratio were determined. The work compares coals in respect of their potential to form NO. On the basis of statistical and regression analysis an attempt to define the relation between fuel properties and NO emission was made. The following conclusions were made on the basis of the conducted research:

- Within one coal type (originating from one mine) the increase of nitrogen oxide emission is observed only to a lesser extent in comparison with the increase of nitrogen content in coal. For the analytical state the nitrogen content increase by 100% resulted in emission variations by some 30%. Such performance of nitrogen oxides emission variations confirms the limited role of nitrogen content in estimating potential emission of nitrogen oxide in coal combustion. Volatile matter content in coal holds a significant part, it is defined on the basis of fuel ratio – FR which is understood as a ratio of fixed carbon content in fuel to volatile matter content.
- 2. It has been observed that the influence of coal type on nitrogen oxides emissions and nitrogen conversion ratio is significant. The emission grows with the increase in rank (carbon content) and it is decreased for anthracite coals. With the increase of nitrogen content the increase of  $NO_x$  emission and decrease of fuel nitrogen conversion ratio are observed. Nitrogen conversion ratio grows with the increase of volatile matter content, it reaches the highest value for brown coal, while nitrogen oxides emissions reaches the highest level for bituminous coal due to a higher content of nitrogen compounds in coal. On the basis of statistical analysis two formulas were determined, they describe variations in nitrogen oxide emission and conversion ratio in stoichometric ratio equal 1.1.

According to the authors of this work research results will have several practical applications. There is a possibility to improve the effectiveness of furnace installation operation by limiting  $NO_x$  emission. Precise control of coal properties should allow to eliminate coals with problematic technical and technological parameters or help to adjust them for use. The classification of particular coals applied in power engineering because of their potential for  $NO_x$  formation will allow to establish a type of catalogue of raw materials. Thus a tool will be created to help to optimise the design and exploitation of new combustion chambers or primary  $NO_x$  reduction measures.

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#### WIESŁAW RYBAK, WIESŁAW FERENS, ARKADIUSZ MACZUGA

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Received: October 9, 2002, accepted: October 2, 2003.