Numerical modeling of nitrogen oxide emission and experimental verification

Lech Szecowka[1] *and* ***Monika Poskart***[1]

Numerické modelovanie emisie oxidu dusíka a experimentálne overenie

The results of nitrogen reduction in combustion process with application of primary method are presented in paper. The reduction of NO_x emission, by the recirculation of combustion gasses, staging of fuel and of air was investigated, and than the reduction of NO_x emission by simultaneous usage of the mentioned above primary method with pulsatory disturbances.

The investigations contain numerical modeling of NO_x reduction and experimental verification of obtained numerical calculation results.

Key words*: numerical modleing, nitrogen oxides, emission reduction of NO_x.*

Introduction

Combustion of fuels is the most important method of primordial energy producing (85 – 95%) and simultaneously it is a main source of toxic components polluted to the atmosphere. About 70% of NO_x and SO_2, 60% of CO, 55% of dust and 98% of CO_2 emission comes from combustion process of fuels. The number of harmful substances created in those processes exceeds 70. That's why processes of fuels combustion should be optimized according to both energetic and ecologic criteria. Energetic criteria require establishing the complex technological parameters (capacity, temperature, pressure, composition of products, lost of heat and others) with the possibility of the highest energy efficiency. Ecological criteria lead to minimization of harmful substations emission.

The pollution can be considered as all substances, which are not present in atmospheric air or present in higher concentration than in pure air. The natural criterion of the pollutants partition is the criterion of the source of its appearance.

There are many technical methods, primary and secondary, of pollutants limitation or reduction, the choice of accepted method is a function of expected results and costs (Wilk, 1998).

Nitrogen oxides are one of most harmful components polluting atmosphere. They are mainly emitted as NO (about 95%), NO_2, and N_2O. Destructive influence of nitrogen oxides on natural environment is commonly known. It is considered that they have a contribution of about 30% in acid rains formation, also in metals corrosions, buildings walls erosion and in damages of forest stand. As a natural background of pure air a value of 0.005 ppm NO_x is accepted. Reduction of emission of gaseous pollution is a strict necessity. It is a result of intensification of combustion processes and the strict standards of environment protection.

This fact forces progress in works of study, introducing new designs of burners and developing special technologies of combustion in order to reduce emission of nitrogen oxides. Up to now more than 70 methods of emission reduction of NO_x have been developed, but many of them are insufficient and very expensive. High investment expenses are particularly the characteristic features of secondary methods. Because of reasons mentioned above new (more effective within a frameworks of methods) solutions are consistently being searched.

The point of those methods is preventing of NO_x formation in industrial flames. Taking into the consideration mechanisms of forming NO_x, methods of emission reducing can be divided into three technological groups: decreasing of high combustion temperature, keeping of local oxygen concentration on a level adequate to excess air 0,7 – 0,9 and special combustion methods (Zelkowski, 1996 and Szecowka, Radomiak et al., 2000).

There is a possibility of increase the reduction efficiency of nitrogen oxides by the use simultaneous a few primary methods of reduction. The most of published scientific reports contains reduction of NO_x by using only single method. Many works were focused on nitrogen oxides reduction. The obtained results are the base for further development of these methods (Slupek, Szecowka et al., 2002; Muzio and Quartucy, 1997 and Zhao, Brereton et al., 1997). In recent years numerical modelling of combusting processes became an uncancellable element of experimental researches. Most of the works deal with both experimental researches and modelling, particularly for comparative analysis. Consequence of this, the series of models of nitrogen oxides emission reduction were created (Jones, Leng, 1996; Li, Williams, 1999 and Fan, Liang et al., 1998). Those models involve primary methods such as fuel and air staging and combustion gasses recirculation. Based on the elements

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of diffusion and heat exchange combined with chemistry of process many of programs such as: FLUENT, KIVA, CHEMKIN and others have been developed. In this paper results of complex researches of simultaneous using several primary methods are presented.

Experimental method

The programme of presented research was as follows:
- initial experimental tests,
- numerical modeling of NO_x reduction,
- experimental verification of obtained numerical calculation results.

The experimental tests were carried out using laboratory stand presented in Figure 1 (Slupek and Szecowka, 2002).

Reactivity combustion chamber was made of three quartz segments 1 m long and 0.12 m in diameter. Separate segments isolated with fibre materials were equipped with measurements holes and nozzles for injecting reburning fuel and second stage air. Main burner was situated at the front of chamber. Natural gas was used as main and reburning fuel. In the middle segment the air under the disturbance pulsation was injected. Mechanical pulsatory generator generated the transient pressure. Frequency of pulsation was controlled by electronic tachometer.

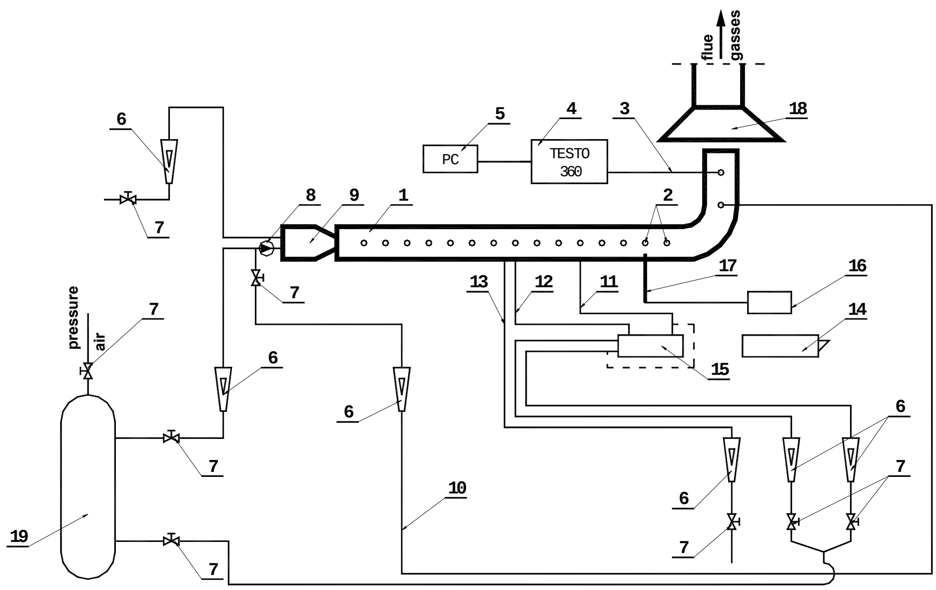

Fig.1. Laboratory for combustion tests: 1 – quartz chamber, 2– measurement points, 3 – probe, 4 – 360 Testo exhaust-gas analyzer, 5 – PC computer, 6–rotameters, 7 – valves, 8 – air injector, 9 – burner, 10 – system of combustion gas recirculation, 11 – system for after-burning air, 12 – system for the secondary air, 13 – system for reburning fuel, 14 – electronic tachometer, 15 – pulsation generator, 16 – measuring card, 17 – PtRh-Pt thermocouple, 18 – combustion gas support, 19 – container of compressed air.

Experimental procedure

All the experimental tests were carried out with the same flow parameters of natural gas and combustion air. The parameters of combustion process as: excess air, NO_x concentration in flame and temperature of outlet combustion gasses were established during the initial tests which was carried out before essential testes.

NO_x concentration was established on the level of 165 ppm, and excess air was equal λ= 1.07. The combustion gasses for recirculation were taken from outlet channel and inject to combustion air in the burner.

The parameters of experimental test were as follows: recirculation step rc = 0.07 – 0.15, volume fraction of reburning fuel rb = 0.05 – 0.17, second stag air to total combustion air ratio rp = 0.11 – 0.26. The frequency of disturbance pulsations in reburning zone was equal to 30 Hz. For all the tests temperature distribution along combustion camber was measured.

Numerical calculations

The results obtained during initial experimental tests were helpful to establish the boundary conditions in the modeling procedure. In the numerical modeling Chemkin programme, version 3. 6.0 has been used. All calculations as a first step of procedure have been done using two applications, Equil and Premix. The model of methane combustion was extended by the reaction of NO_x formation. All the reactions and the equilibrium constants have been taken from the Miller – Bowman model.

The model presented in this paper contained 126 reactions and 28 chemical compounds. The next step of procedure was to prepare input file contains the conditions of experiment: mass flux, pressure, temperatures profile and proper calculation grid.

The following quantities have been used:

- mass flux 2.8 – 3.9 g/m^2s,
- the temperatures profile taken from the initial experimental tests,
- the substrates calculated by Equil as a fraction rate,
- estimated products of combustion calculated by Equil as a fraction rate.

In the next stage of investigations, the verification of the results was carried out by numerical simulations and experimental measurements.

First, the reduction of NO_x emission, by the recirculation of combustion gasses, staging of fuel and of air (used separately) was investigated, and than the reduction of NO_x emission by simultaneous usage of the primary method:

- recirculation of the combustion gasses, reburning and pulsatory disturbances,
- staging of air, reburning and influence pulsatory disturbances,
- staging of air, recirculation of the combustion gasses and pulsatory disturbance was discussed.

The pulsatory disturbances were generating in the middle segment of the chamber. It intensifies the mixing of second stage air and reburning fuel. During the experiments, the distribution of the temperature and the NO_x concentration along combustion chamber and NO_x concentration at the end of chamber for all tests have been measured. The results of measured and calculated parameters are shown in Figures 2 – 8.

Analysis of results

Numerical modelling of the reduction of NOx emissions by fuel staging, showed the increase of the process efficiency with the increase of reburning fuel ratio. The experimental tests showed the numerical results and measurements (Fig. 2). A good agreement between the numerical modeling and experimental results was obtained for the simultaneous applying the recirculation of combustion gasses and the reburning (Fig. 3).

It is essential to point out that the efficiency of the process resulting from the numerical calculation was always higher then in the experiment because of assumptions involved in the model ideal mixing of reacting substances. The good consistence of the results of assumed model and verification tests permit to extension experiments range.

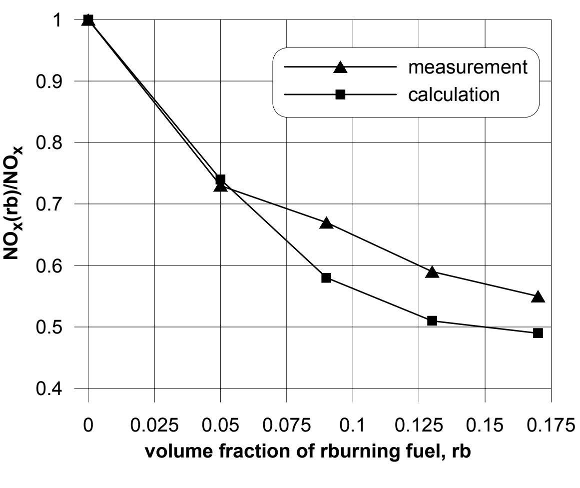

Fig.2. Influence of reburning (rb) method on NO_x reduction.

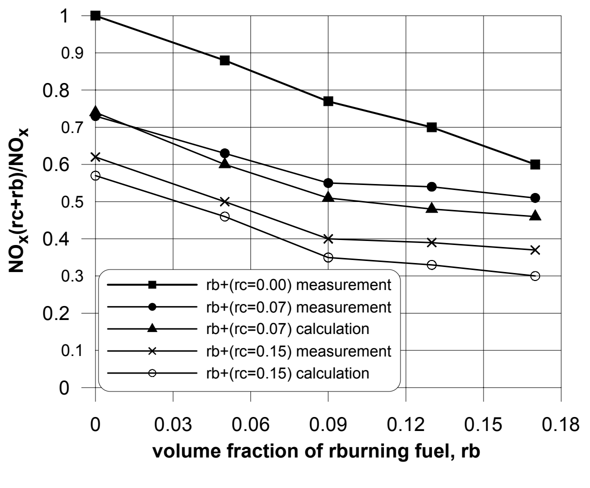

Fig.3. Influence of reburning and recirculation (rc) methods on NO_x reduction.

$$\frac{NO_x\,(rb+rc)}{NO_x} = f(rc)$$

The application of low temperature recirculation and reburning changed the temperature profile in the chamber (Fig. 4) in the highest temperatures zone and a little growth in the middle part of the chamber.

The application of the above methods significantly decline of NO_x concentration along the chamber. The injection of the reburning fuel caused further decline of NO_x concentration (Fig. 4 and 5). The pulsatory disturbances additionally intensified the process of NO_x reduction. The final effect of NO_x reduction was up to 71%. The results of the next tests when the simultaneous, the air staging and reburning methods were applied are shown in figures 5 - 7. The temperature (Fig. 5) indicates its significantly decrease up to the middle of the chamber. It strongly effects the additional decline NO_x concentration in combustion gases (Fig. 6 and 7), but the influence of air staging was greater than reburning. The final result of the NO_x reduction was equal about 89%. The next experimental tests included simultaneous application of air staging and combustion gases recirculation (Fig. 8). The air staging was the primary method. The additional application of combustion gases recirculation caused even worse effects than using only air staging.

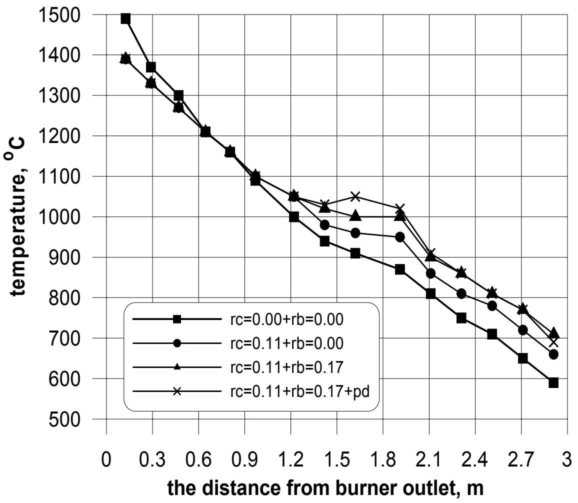

Fig.4. The distribution of temperature along the combustion chamber during recirculation, reburning and pulsation disturbances.

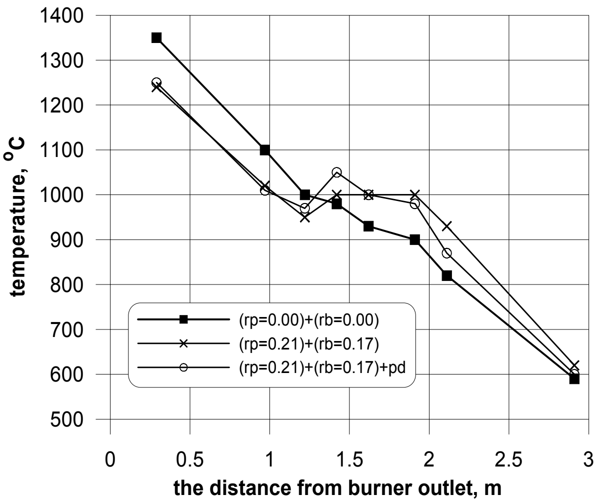

Fig.5. The distribution of temperature along the combustion chamber during air staging, reburning and pulsation disturbances.

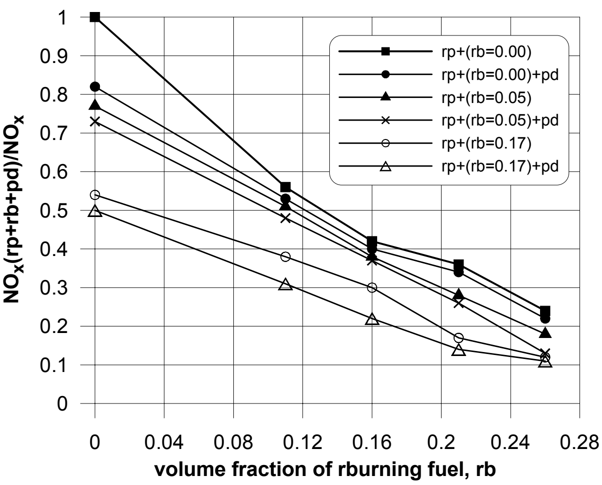

Fig.6. The impact of the air staging and reburning on reduction NO_x concentration

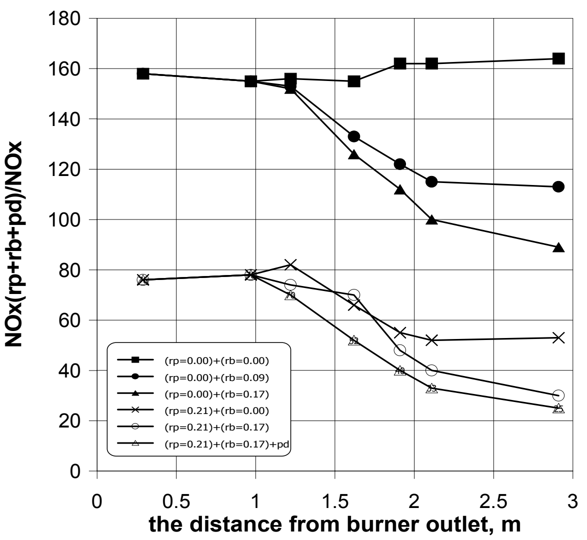

Fig.7. The distribution of NO_x concentration along the combustion chamber during air staging, reburning and pulsation disturbances.

$$\frac{NO_x\,(rp+rb+pd)}{NO} = f(rp)$$

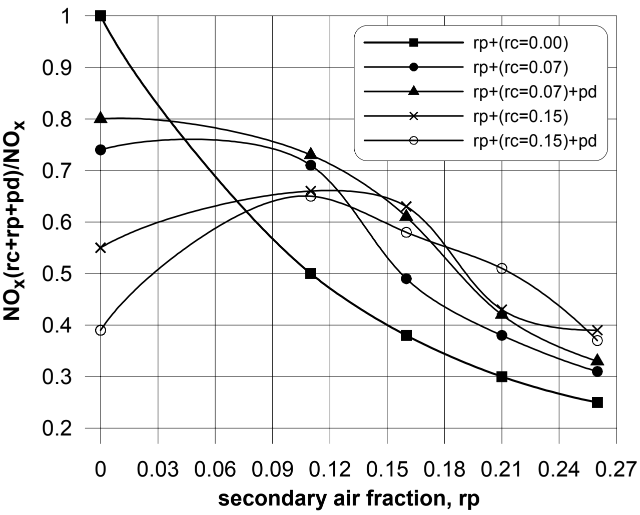

Fig. 8. The influence of the air staging, recirculation and pulsation disturbance on reduction in NO_x concentration

$$\frac{NO_x\,(rc + rp + pd)}{NO_x} = f(r_p)$$

Conclusions

The numerical calculations and experimental tests showed that simultaneous application of primary methods cause additional increase of efficiency of NO_x concentration reduction. The greatest efficiency of NO_x reduction in combustion processes of natural gas has been obtained applying the air staging and reburning, supported by pulsatory disturbance of the reagents.

The increase of the NO_x reduction of 14% in comparison to fuel staging, and of 44% in comparison to reburning has been observed. This effect is a consequence of a specific character of both methods.

The simultaneous application of recirculation and reburning appeared to be very efficient. It caused the 25% increase of NO_x reduction. The combining air staging and combustion gases recirculation does not reduce the NOx emission therefore cannot be recommended.

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