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The application of RANS CFD for design of SNCR technology for a pulverized coal-fired boiler

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The article describes the technology of NO_x emission abatement by SNCR method. The scope of research included CDF simulations as well as design and construction of the pilot plant and tests of NO_x reduction by urea in the plant located in industrial pulverized-coal fired boiler. The key step of research was to determine the appropriate temperature window for the SNCR process. The proposed solution of the location of injection lances in the combustion chamber enabled to achieve over a 30% reduction of NO_x. It is possible to achieve higher effectiveness of the proposed SNCR technology and meet the required emission standards via providing prior reduction of NO_x to the level of 350 mg/um^3 using the primary methods.

Keywords: NO_x emission abatement, power plant boilers, selective non-catalytic reduction, flue gas treatment.

INTRODUCTION

The emission of nitrogen oxides, NO and NO_2 (commonly labeled as NO_x), formed as by-products in the processes of fuels combustion, is an increasing problem for the Polish energy sector. Due to the scale of production, this problem particularly affects the coal-fired power plants. The process of coal combustion in boilers is accompanied by the formation of nitrogen oxides in three reaction pathways: *thermal*, *fuel* and *prompt*. The contribution of NO_x, produced by the respective mechanism depends on the combustion conditions (flame temperature, residence time in the combustion zone, the excess oxygen used in the combustion process and the amount of nitrogen in fuel)[1].

Since the nineties of the last century, due to the strong negative impact of nitrogen oxides on the environment and living organisms, the legislative regulations, governing the permissible NO_x emission level in energy, transport and chemical industry and waste disposal have been created and amended. At the beginning of 2016, the *Industrial Emissions Directive* (*IED*) which imposes an obligation for significant reduction of nitrogen oxides emission on the EU countries came into force[2]. The NO_x emission standards for coal-fired power plants with a nominal capacity of more than 100 MW determine the maximum permitted NO_x emission at a level of 200 mg/um^3 (referred to the content of NO_2)[3].

These more stringent standards regarding the NO_x emission, force the companies from the industrial sector to use technologies which would enable them to reduce the nitrogen oxides content in the exhaust gases. In the existing coal-fired boilers, it is preferred to use the primary methods which rely on the limiting of NO_x formation at the stage of fuel combustion. The implementation of these methods is related to the modification of combustion process via the use of specially designed burners (Low NO_x Burners). Although the primary methods require the interference in the combustion process or their individual adaptation to the existing boilers, they are widely used due to the low cost of their implementation.

Unfortunately, the use of only primary methods is not sufficient to fulfil more and more stringent emission requirements. Therefore, it is also necessary to apply secondary methods which rely on the reduction of nitrogen oxides formed in the combustion zone. The secondary methods are divided into catalytic (SCR) and non-catalytic selective reduction (SNCR).

The SCR method is based on the nitrogen oxides reduction in the flue gases by using a catalyst and a suitable reductant (the most commonly used reducing agent is ammonia). The application of the catalyst allows for carrying out the process with high efficiency (up to 95%) at the low temperature (150–425°C)[4]. The disadvantage of the SCR technology is its high capital cost. Its application in existing plants requires the modification of this part of the plant where the temperature is low and the installaton of the SCR reactor with a suitable catalyst. The requirements vital to achieve high NO_x reduction efficiency by the SCR method include: cleaning the flue gases from the acidic contaminants and ashes, preheating the flue gases to the suitable temperature (additional equipment and higher energy consumption – higher operating costs) and the application of the catalyst (which should be replaced from time to time)[5].

There are many types of catalysts for the SCR process. The most commonly used are vanadium-based (e.g. V_2O_5/TiO_2) and doped-zeolite catalysts[6] which in contrast to very active precious-metal bearing catalysts (e.g. platinum, platinum stabilized with barium) exhibit a better selectivity in a wider temperature range[6]. Other significant advantages, related to the use of vanadium-based systems are lower cost of the catalyst and the resistance to sulphur compounds which are often present in the exhaust gases.

Due to the high costs of SCR method, the attractive alternative is the SNCR technology. The main advantage of *Selective Non-Catalytic Reduction* method is the possibility of its implementation in existing boilers without significant modifications in industrial plant, as well as much lower capital and operating costs. The SNCR technology relies on NO_x reduction in the flue gases stream to nitrogen, water vapour and carbon dioxide. The reducing agent can be ammonia (gaseous or aqueous solution), urea (solid or aqueous solution) or cyanuric acid. Usually, a few aspects should be taken into account when selecting the reductant for the SNCR process: capital and operating costs, physical properties of the reductants as well as operational considerations and safety reasons.

Urea is a nontoxic and less volatile liquid that can be stored and handled more safely than ammonia. Urea solutions have better dispersion properties after injection into the boiler and therefore their droplets can penetrate farther into the flue gas than ammonia what enhances the degree of urea spray mixing with the flue gas. For this reason, urea is typically used in the SNCR process in large boilers where good mixing is difficult to achieve. Urea-based injection systems typically have a modular design which can easily meet the requirements of the boiler-specific design and this facilitates the construction of these systems on the boiler (this minimizes the capital costs)[7, 8].

In the case of aqueous solution, the process of urea transformation into amino radicals occurs gradually. At the first stage of SNCR process, the water evaporates from the reductant droplets and then urea is decomposed[9]. It provides a better penetration of amino radicals inside the combustion chamber and their better mixing with the flue gas.

In general, the SNCR process can be described by the equation (1):

$$CO(NH_2)_2 + 2\ NO + \tfrac{1}{2}\ O_2 \Leftrightarrow 2\ N_2 + CO_2 + 2\ H_2O \quad (1)$$

At the temperatures above 300°C, the reaction of urea decomposition occurs in the following way:

$$CO(NH_2)_2 \rightarrow NH_3 + HNCO \quad (2)$$

The reaction products combine with water vapour forming NH_2^*, H^* and OH^*radicals which then react with nitrogen oxides according to the equations (3)–(5):

$$NH_2^* + NO \Leftrightarrow N_2 + H^* + OH^* \quad (3)$$

$$4\ NH_2^* + 2\ NO_2 \Leftrightarrow 3\ N_2 + 4\ H^* + 4\ OH^* \quad (4)$$

$$HNCO + NO + H^* \Leftrightarrow N_2 + CO + H_2O \quad (5)$$

An important technological condition for the effectiveness of SNCR method is to find a way for uniform distribution of the reductant in combustion chamber at a suitable temperature zone, so called "temperature window" of SNCR process. The satisfactory reduction levels can be achieved in the temperature range from 850 to 1050°C[9]. When injecting the reducing agent into the zone of too low temperature (usually $T<850°C$), the effect of *ammonia slip* may occur resulting in the appearance of NH_3 in exhaust gases or in dust. At the temperature above 1050°C, the reactions leading to the formation of additional amount of NO_x that can decrease the reduction effectiveness or even increase the concentration of nitrogen oxides in the exhaust gases dominate. In practice, the effectiveness of the SNCR method does not exceed 50%.

A Selective Non-Catalytic Reduction in a pulverized coal – fired boilers occurs under conditions of turbulent gas flow which causes the fluctuations of velocity, temperature and composition of the flue gas inside the combustion chamber. When estimating the temperature zone proper for the SNCR technology, simplified CFD RANS simulations should be sufficient to obtain the information about the spatial distribution of the temperature inside the boiler. However, we should bear in mind the fact that the industrial SNCR process operates itself with flows at high Reynolds numbers and it is characterized by multiple reductant injections at various enthalpies and chemical compositions. A turbulent mixing of the reductant spray with the flue gas and evaporation of its droplets can result in temporal and local fluctuations of the temperature in deNO_x zone, making it difficult to ensure this temperature within the proper range. Thus, in order to obtain the precise information about the flow conditions in the reducing zone, not only the average temperature values at chosen reduction levels should be taken into account, but also the temperature fluctuations arising from the topology of the spray[10, 11]. According to Farcy et al.[10], the temperature fluctuations can have quite a significant influence on the efficiency of NO_x reduction, causing drop of the reduction degree even by several tens of percent in the zones of high temperature fluctuations when using ammonia as a reductant. In order to accurately analyze the influence of temperature fluctuations on SNCR process, RANS calculations should be complemented with large eddy simulation LES. These simulations may be used to optimize the geometry of the injection lances.

An experimental determination of the injection sites of reductant is complicated due to the methodological difficulties associated with the temperature measurements, boiler dimensions and a high dynamics of the operating parameters changes. The temperature fluctuations in the cross-sectional area of the combustion chamber can reach several tens degrees Celsius. For this reason, the experimental measurements lead to the estimated values, which should be confirmed for example, by the above mentioned numerical calculations.

RANS CFD (Reynolds-Averaged Navier-Stokes Computational Fluid Dynamics) numerical analysis was used to design the SNCR plant for a pulverized coal-fired boiler and determine the temperature window of the process.

EXPERIMENTAL

The research work was divided into the following stages: numerical calculations (CFD), design and construction of the plant, located on the industrial pulverized coal-fired boiler, and SNCR tests.

In order to find the temperature zone suitable for the SNCR process, the numerical CFD calculations were performed. They included determination of the temperature and flue gas velocity profile and specification of distribution of NO_x concentration and the residence time of urea solution droplets in the injection zone for different operating conditions of the boiler.

The computational domain was based on the conceptual design, containing burners arrangements and air inlets location (Fig. 1). The arrows indicate the location of dust burners, secondary air inlets from the side of boiler throat and flue gas outlet after the 1-st stage steam superheater. In the numerical simulation, the flue gas flow through the combustion chamber and steam superheaters zone was included. The two subdomains, representing steam superheaters zone were separated from the main

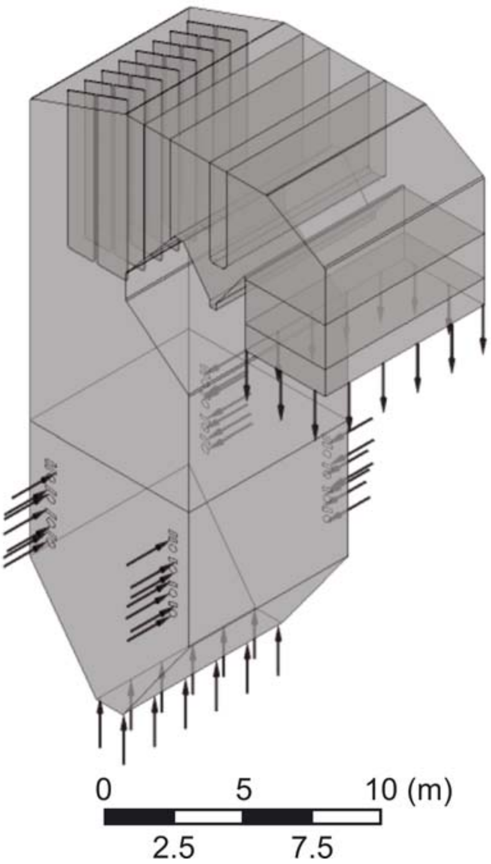

Figure 1. The geometry of calculation domain

computational domain. The 1-st and 3-rd stage steam superheaters were treated in CFD simulations as one calculation block. The 2-nd stage steam superheater was modelled as two calculation blocks (vertical and horizontal), because of the large distances between the consecutive pipe layers. The flue gas flow through the part of superheater piping was simulated at several gas velocities for each subdomain. Based on these results, the dependence of the pressure loss on the gas velocity, permeability and the pressure loss coefficient were determined for the dominant gas flow direction.

Simulations and calculations were performed for the steady state conditions and they included the reaction of pulverized coal combustion, the flue gas flow and convective and radiative heat transfer.

The combustion of pulverized coal in boiler consists of three main stages: I-coal degassing, II – combustion of volatile compounds and III – char combustion. The first process is a multiphase reaction and it represents the phenomenon of the gaseous substances releasing from the coal fuel. The products of this reaction are Gas Fuel (gaseous state) and Char (solid state). The devolatilization process of the coal was described as a single first order reaction. Moreover, it was assumed that Gas Fuel is oxidized to CO_2 and H_2O and the Char is oxidized to CO_2.

The mechanism of nitrogen oxides formation was based on two models: the thermal and prompt one. The model of droplets evaporation used in calculation included heat and mass transfer processes between droplets and the surrounding area. This model used mass transfer equations for the conditions corresponding to the temperatures below and above the boiling point.

Simulations were performed for different boiler loads and for different amounts of urea solution introduced into the combustion chamber.

The investigations of selective non-catalytic NO_x reduction with the use of urea solution as a reducing agent were performed in the plant located at the pulverized coal-fired boiler with a nominal heat output of 171 MW. The plant consists mainly of: reductant dosing station and the system for injecting urea solution into the boiler. Its project assumed the minimal interference in the steam-heating system of the boiler. This entailed the necessity of using the injection lances of the external diameter not exceeding 30 mm.

The method of injection lances installation in the boiler wall is shown in the photo (Fig. 2).

The implemented proprietary design of the reductant injection system ensures a trouble-free injection of a reductant in the high temperature zone and it provides the protection against overheating and potential melting of the lances or nozzles responsible for the proper distribution of the reductant in the combustion chamber. The assembly of this system was approved by the Technical Inspection Office[12].

The SNCR investigations were carried out in the flow of a real flue gas mixture produced by the combustion of pulverized coal at the stechiometric excess of the combustion air, measured by the concentration of oxygen in the exhaust gases (in the range of 5.5–7.5% vol.). Tests were performed in a typical technological regime of the boiler operation at the boiler loads of 70–100% of nominal boiler capacity. The urea solution with the concentration of 6–23% wt. was injected into the boiler at the rate of 300–500 l/h, through the lances installed at two boiler levels: 11 lances on the lower and 9 lances

Figure 2. Photo of an injection lance installed in the boiler wall

on the upper level. During the test, the bolier load and the concentration of O_2 and NO_x in the exhaust gases were registered. The NO_x concentration in flue gas was referenced to 6% vol. of O_2 in the exhaust gases (mg/um^3).

The degree of NO_x reduction ($C_{NOx,\ measured}$) was determined on the basis of NO_x concentration measured during injection of urea into the boiler with its reference to the concentration of NO_x formed during coal combustion process and measured in the absence of the reductant ($C_{0,NOx}$) according to the following equation:

$$Red_{(NOx)} = \frac{C_{0,NO_x} - C_{NO_x measured}}{C_{0,NO_x}} \cdot 100\% \qquad (6)$$

where:

$C_{NOx\ measured}$ – the average concentration of NO_x measured during urea injection, mg/um^3

$C_{0,\ NOx}$ – the concentration of NO_x measured in the absence of the reductant, mg/um^3

The degree of nitrogen oxides reduction was determined as an average NO_x concentration registered during injection of the fixed amount of urea solution into the boiler for at least 45 minutes. In order to achieve the uniform distribution of urea solution in the combustion chamber, flat nozzles with different injection angles were used (in the range of 45–120°), depending on the location of lances in the chamber. The NO_x concentration was measured using on-line analyzer which is the part of the boiler equipment.

RESULTS AND DISCUSSION

The results of numerical calculations (CFD) performed for pulverized coal combustion process, indicate the significant differentiation of the flue gas temperature and NO_x concentration in the entire boiler volume and the occurrence of turbulences in the flowing flue gas (Fig. 3).

The analysis of the boiler operating parameters and CFD calculations enabled to determine the zone of the optimal "temperature window" for NO_x reduction which is located at the height equal to a minimum 19.0 m. The upper height of injection zone is in practice limited by the boiler construction (the presence of the 3-rd stage steam superheater). Moreover, the boiler construction allowed for the installation of injection lances only on three walls of the combustion chamber (instead of four). On the basis of the results of *CFD* simulation, the decision about the location of injection lances at two boiler levels was made. This solution allows for a greater flexibility of the reductant injection. Depending on the current operating parameters, it is possible to inject the urea solution at one boiler level or at two levels at the same time. The location of the upper lances row was set at the height that prevents direct acting of the urea solution on the elements of the 3-rd stage steam superheater. The schematic layout of the injection lances at the lower and upper boiler level is shown in Figure 4.

The analysis of the isothermal surface in the upper area of the "temperature window" (1050°C, Fig. 5) shows that at the considered level of urea injection, a zone with too high temperature for the SNCR process occurs (the isotherm interior). Therefore, the injection of reductant into a greater depth of the boiler can lead to the formation of additional amount of NO_x, due to too high temperature which prevails locally there.

At the temperatures exceeding the upper limit of the "temperature window" for urea-SNCR, the fast oxidation of ammonia to NO processes are activated. Under these conditions, the concentration of OH radicals starts to build up and initiate NH formation:

$$NH_3 + OH \leftrightarrow NH_2 + H_2O \qquad (7)$$

$$NH_2 + OH \leftrightarrow NH + H_2O \qquad (8)$$

Once NH is formed, the sequence of high temperature oxidation reactions lead directly to nitric oxide formation:

$$NH + O_2 \leftrightarrow HNO + O \qquad (9)$$

$$HNO + O \leftrightarrow NO + OH \qquad (10)$$

At sufficiently high temperature, the oxidation sequences become significantly dominant and they may result in a net increase in the NO concentration.

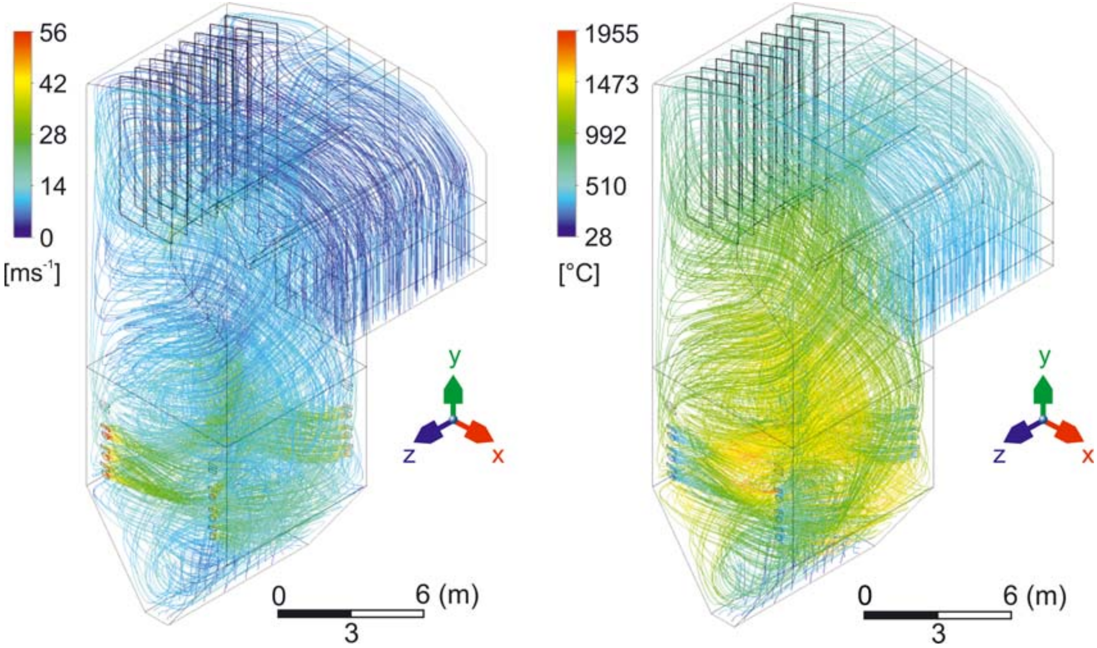

Figure 3. The results of numerical calculations (CFD): flue gas velocity profile (on the left) and the temperature profile (on the right side) on the streamlines in the pulverized coal-fired boiler

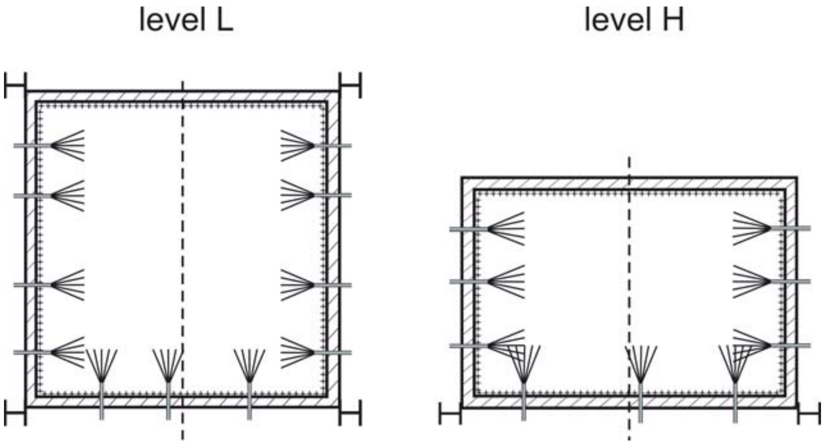

Figure 4. The diagram of injection lances location in the pulverized coal-fired boiler

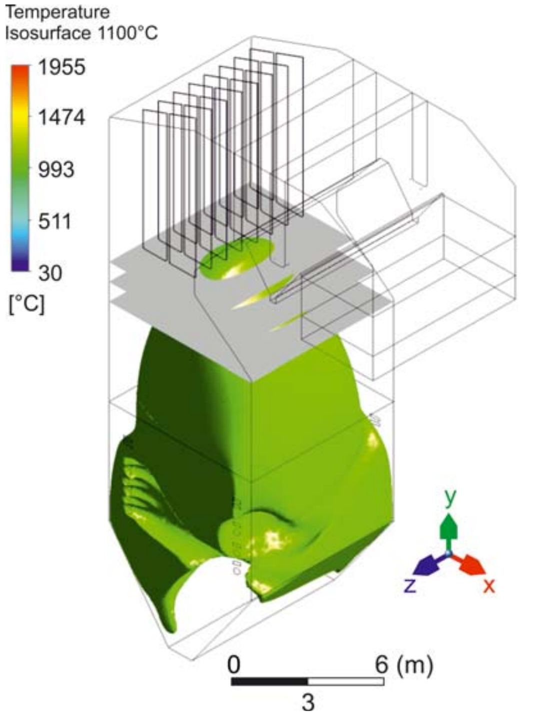

Figure 5. The isothermal surface calculated for the temperature of 1050°C

On the basis of *CFD* simulations and with regard to the injection process of urea solution, the sizes of nozzles orifices were selected. The chosen solution enabled to limit the range of reductant injection to the depth of 2.5 m from the boiler wall. The results of *CFD* simulations (Fig. 6), indicate that all reductant droplets evaporate (droplets diameter is reduced to 5% of initial diameter) within 0.15 s after injecting the urea solution into the combustion chamber. It means that droplets evaporate almost completely before reaching the zone, having a temperature too high for the SNCR process. Only a few reductant droplets, injected through the nozzles from the lower row of lances (particularly at a high boiler load), can enter this zone.

The results of *CFD* simulations presented above were verified experimentally in the SNCR plant, mounted on the industrial boiler. The measurements of NO_x reduction effectiveness, $Red_{(NOx)}$ were performed at various boiler capacity *BC* [%] and different stechiometric excess of oxygen $C_{NOx,meas.}$ for combustion process, measured by the concentration of oxygen at the outlet $C_{O2,comb.}$ [% vol.]. These parameters determined also the initial concentration of NO_x in the flue gas $C_{0,NOx}$ [mg/um^3]. Investigations were performed for two flow rates of urea solution F_u [l/h] having a concentration in the range of 6–23% wt. C_u, by injecting the reductant solution onto the one selected level (H or L) or two levels simultaneously. The results of the tests are summarized in Table 1.

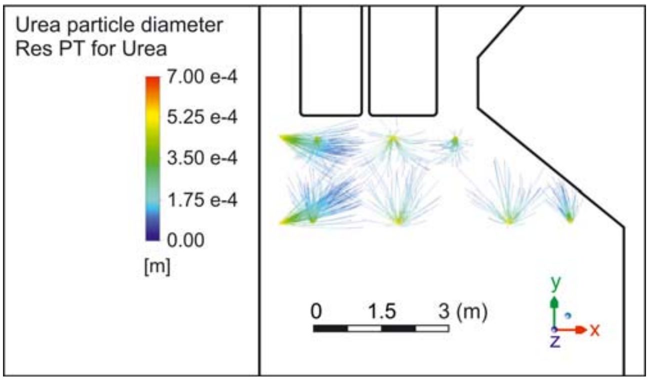

Figure 6. The results of numerical calculations related to the evaporation process of reductant droplets

The process of NO_x reduction occurred during the injection of urea solution at one selected boiler level or at two levels simultaneously. This proves the appropriate choice of zone for injecting the reductant. The efficiency of NO_x reduction is influenced by the operating conditions of boiler, mainly by load and the amount of excess air for combustion. The higher the concentration of urea solution, the higher the degree of NO_x reduction is but it carries a potential risk of ammonia slip in the exhaust gases. Therefore, the use of higher flow rates of dilute urea solution is preferred for NO_x reduction process. Thus, the concentration of the injected solution should not exceed 15% wt. The average NO_x reduction observed during SNCR tests was at the level of 30%, but at lower boiler loads it reached even more than 45%.

One of the conditions for an effective NO_x reduction is the uniform distribution of the reductant solution in the entire reaction zone with a suitable temperature. The proposed solution, based on a proper selection of the nozzles with different injection angles depending on the location of nozzles on the boiler walls brought the expected results. The best results were obtained by installation of the nozzles with larger injection angles in the central part of the boiler wall and with smaller injection angles at the corners of the combustion chamber. The results shown in Table 1 relate to SNCR tests carried out using the proposed configuration of the nozzles.

Table 1. The results of investigations of the selective non-catalytic NO_x reduction

Operating parameters of the boiler			Urea solution			$C_{NOx,meas.}$ [mg/um^3]	$Red_{(NOx)}$, [%]
BC, [%]	$C_{O2,comb.}$ [% vol.]	$C_{0,\ NOx,}$ [mg/um^3]	C_u, [% wag.]	F_u, [l/h]	Injection level		
85	5.6	400	15	300	L	293	26.9
88	7.1	540	11	500	H	415	23.1
92	5.6	420	15	300	H	286	31.9
74	6.2	350	23	500	H	189	45.9
91	7.0	550	6	500	H	443	19.5
79	5.9	400	11	500	L + H	287	28.2
84	5.6	400	15	500	L + H	273	31.9
80	6.3	400	23	500	L+H	269	32.7

NO_x concentration was reffered to 6% vol. of oxygen concentration in the exhaust gases.

Due to the level of final NO_x emission, it is preferred to use the lowest possible excess of the combustion air. The modification of the flue gas composition and the temperature profile in the established *"temperature window"* zone can lead to obtain the low primary NO_x emission level by optimizing the amount of air and the method of its distribution. It is correlated with the optimization of the combustion process (primary methods) which is beyond the scope of the presented research.

The analyses of the flue gas composition leaving the SNCR zone and flowing to the chimney, as well as analyses of ashes composition for the ammonia content indicate that the SNCR process does not lead to the increase of ammonia concentration in exhaust gases or ashes.

CONCLUSIONS

The numerical analyses of the urea droplets residence time in the combustion chamber were performed for the selected optimal injection zones and nozzles. The investigations of NO_x reduction by SNCR method were carried out in the plant located at the pulverized coal-fired boiler with a nominal capacity of 171 MW and under actual operating conditions of the boiler in the flow of untreated flue gases. On the basis of these results, working conditions of the plant for the urea solution injection into the boiler were pre-determined. These conditions enabled to achieve over a 30% NO_x reduction. The SNCR tests have shown that it is possible to achieve low NO_x emission (below 200 mg/um^3) for the proposed location and arrangement of the lances providing its prior reduction to the level of 350 mg/um^3 via primary methods.

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