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Effects of operating conditions on deNO$_x$ system efficiency in supercritical circulating fluidized bed boiler

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Abstract

The purpose of this work was to determine the impact of a operating conditions on the deNO$_x$ system efficiency.in a 966 MW$_{th}$ supercritical circulating fluidized bed boiler. Experimental tests were carried out on a full-scale DeNO$_x$ system installed in the world's largest once through supercritical circulating fluidized bed boiler. In this work, the effects of the following parameters were studied: flue gas temperature inside the separators between 636°C and 845°C, relative ammonia mass flow over the range 0.22–1.00 and three relative values of O_2 concentration (i.e. 0.94, 1.0 and 1.13). The efficiency of deNO$_x$ system increases (ca. 53%) with increasing relative ammonia mass flow. A maximum DeNO$_x$ system efficiency (ca. 70%) was achieved at flue gas temperature in the range from 720°C to 790°C. In the case of all unit loads, deNO$_x$ system efficiency from 36% to 70% was observed and performs a standard emissions relate to permissible concentration of NO$_x$ in the flue gas.

Keywords: Supercritical CFB boiler, SNCR method, deNO$_x$ system efficiency, Operating conditions.

1. Introduction

Recently, circulating fluidized bed (CFB) combustors are included to major direction of retrofit power engineering in Poland. Those activities influence on improve efficiency of power units and reduction of air pollutant emission from coal fired CFB boilers. Nitrogen oxides (NO$_x$) is the main product of fossil fuel combustion. The chemical symbol NO$_x$ collectively represents two different compounds i.e. nitric oxide (NO) and nitrogen dioxide (NO_2). NO is colourless and an odour-free gas, whereas NO_2 is a pungent reddish-brown gas. The NO$_x$ emissions to the atmosphere from fossil fuel-fired boilers consist of 95% NO and the reminder less than 5% NO_2 [1, 2]. NO$_x$ emissions in CFB facility have been investigated by many authors and can be found in [3–12].

There are three principal forms of NOx: thermal NO$_x$, fuel NO$_x$ and prompt NO$_x$. Thermal NO$_x$ formation is the result of reaction of atmospheric nitrogen and oxygen in post-flame region of the furnace at the high level of temperature. The formation of thermal NO$_x$ in CFB combustion processes mainly depend upon following factors: temperature, the concentration of primarily nitrogen and oxygen in the combustion gases and residence time within

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combustion chamber. In the case of CFB technology, the oxygen concentration at the bottom part of combustion chamber equals an average 12.5% and the residance time of the flue gases within furnace of CFB boiler is in the range from 3 to 4s. During solid fuels combustion in fluidized bed boilers, the share of thermal NO_x in nitrogen oxides emissions is less than 10%, because combustion temperature rarely exceeds 1250 K. The reaction mechanism of thermal NO_x has been detailed descriebed by Zeldovich [13], others authors [1, 14, 15] and will not be repeated in this work.

In this work, the objective of the study was an assessment of effect of operating conditions on $deNO_x$ system efficiency. Measurements were conducted for four different unit loads, when the boiler was operated under a steady state. All test runs were performed on 1296 t/h supercritical circulating fluidized bed boiler, fueled with bituminous coal. Reduction of NO_x emission during combustion process based on ammonia water injection into the upper part of the solids separator. The multi-pollutant control during combustion process with deNOx system was not considered in the present study.

2. Review of $deNO_x$ methods for CFB technology

There are two main control methods for reduce NO_x emissions: (i) combustion controls, and (ii) post-combustion controls (i.e. flue gas treatment). Combustion controls limit the amount of NO_x which is generated during combustion process in the boiler. This way to reduce NO_x emission is commonly applied in power and heat generation industry. There are any possibilities of combustion controls by means of following factors: lowering of combustion temperature, air staging, and lowering excess air. In a CFB boilers, the combustion temperature is in the range from 750° to 900°C and allows ensured NO_x emissions well below the permissible emission limit values. At the low combustion temperature, the formation of thermal NO_x is inhibited, however, almost all of fuel-N is converted to NO. The quantity of fuel NO_x proportionally decreases with temperature. Staging of combustion air may by used at the combustion of a wide range of different solid fuels in the CFB boilers. The staging of the combustion air influence on NO_x formation by two ways [16]: (i) the staged combustion results in a cooler flame and (ii) the staged combustion results in less oxygen reacting with fuel molecules. Staging air system consists of a fuel-rich primary zone and fuel-lean secondary zone. As a result of air staging fuel-bound nitrogen is converted to molecular nitrogen in the primary zone. However, in the primary combustion zone are reduced: flame temperature, O_2 level and residence time at peak combustion temperature. The reduction of NO_x emissions by means of typically staged air burners is possible about 30–60% compared to a single-stage injection. The degree of the staging air is limited by a size of loss of incomplete combustion. More descriptions on staging air for laboratory- and bench-scale in fluidized bed combustion are presented in the works [17–19]. The easiest way to reduce NO_x emissions is lowering excess air to the minimum value that allows the complete combustion of fuel. In the case of a commercial CFB boilers is possible reduction of NO_x emission about 50% at the lowering excess air ratio about 20%.

Table 1: The optimum operating temperature for $deNO_x$ catalyst types [20]

Category	Material	Applicable temperature, °C
Precious Metal	Platinum	240–290
Base Metal	Titanium Oxide/Vanadium	245–400
Zeolite	Aliminium Silicates	400–600

Post-combustion $deNO_x$ methods reduce NO_x concentration in the flue gas from the boiler. In a CFB boiler, the most popular and feasible $deNO_x$ methods with an application are dry systems based on injection of chemical agents (i.e. ammonia, urea) to the hot exhaus gases. Dry systems include two main methods of flue gas treatment such as: selective catalytic reduction (SCR) or selective non-catalytic reduction (SNCR).

Selective catalytic reduction a significant reduction of NO_x, generally up to 80–90%. In the case of selective catalytic reduction $deNO_x$ system a treatment of flue gas is carrried out with ammonia within a heterogeneous catalytic bed. The most popular several catalyst types used in SCR method are shown in Table 1. The catalyst in form of metal plate extruded as a honeycomb monolith sections is installed in vertical downward flow between economizer and air heater. The efficiency of SCR deNO$_x$ system depends on flue gas temperature, NH_3/NO_x ratio, oxygen concentration within combustion chamber, contentration of other flue gas constituents, uniform flue gas flow distribution and type of catalyst support used.

Selective non-catalic reduction is a “thermal deNO$_x$” process in which NO_x is reduced to N_2 and water by reaction with chemical agent such as ammonia (NH_3) or urea $CO(NH_2)_2$. In the practice, ammonia or urea is directly normally injected into upper part of the boiler to hot flue gas due to fact that the the optimum temperature for conversion NO_x to N_2 and H_2O is within 870–1200°C [21]. Flue gas temperature greatly affect SNCR system efficiency. Whereas the flue gas temperature is too high, ammonia will be oxidized to NO_x and ultimately to will reduce the efficiency of SNCR system. If the flue gas temperature is to low, the NO_x removal process diminish and unreacted ammonia concentration increase in exhaust gases. There are several important factors which affect performance of SNCR system: proper flue gas temperature, sufficient residence time of the chemical agent and flue gas, degree mixing of ammonia/urea and flue gases, chemical agent-to-NO_x ratio, fuel sulphur content and NO_x concentration in the flue gas. SNCR deNO$_x$ systems have been allowed to achieve chemical NO_x reduction efficiency of approximately 25–60%. The application of SNCR to CFB boilers is technically feasible because the normal operating temperature of CFB is near the optimum temperature window for NO_x reduction by means of ammonia [16]. In the case of amine-based reagents is needed the higher flue gas temperature unlike SCR method.

3. Description of the supercritical CFB boiler

A schematic of the supercritical CFB facility is shown in Figure 1. The CFB unit is designed to generate 1,296 t/h of steam, at 27.5 MPa and 560°C with

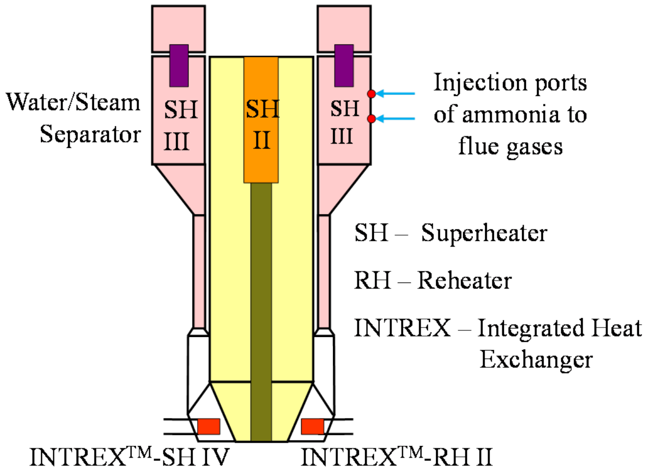

Figure 1: Schematic layout of utility 1296 t/h supercritical CFB boiler at Lagisza Power Plant

feedwater temperature of 290°C. Unlike typical second generation circulating fluidized bed combustion systems, CFB unit is equipped with a vertically tubed BENSON evaporator, which is a new supercritical steam technology. Detailed information with respect to the supercritical steam generation technology is precisely described by [22].

Besides, the boiler consist of the following systems: (i) furnace chamber; (ii) solids separators; (iii) INTREX™ integrated heat exchangers; (iv) low temperature flue gas heat recovery system. The furnace cross section dimensions are 27.6 m×10.6 m, deep and wide respectively. The supercritical CFB boiler has a height 48m and a thermal capacity of 966 MW$_{th}$. The bottom part up to 9.0m height, the walls are covered with refractory lining. On the bottom of combustion chamber is a fluidization grid with nozzles of primary air. Secondary air nozzles are at three levels above the grid in the two sidewalls.

The supercritical CFB boiler is equipped with eight solids separators, four along both sidewalls. Separators are made of membrane walls, which are covered with a thin layer of erosion-resistant refractory. The solids return legs leading to the INTREX™ chambers integrated with the furnace sidewalls have a similar refractory lined design. In the INTREX™ chambers the heat is transferred from circulating material to the final superheater and reheater stages. In the sloped section of the right and left walls, there are openings for bed material return from the

Table 2: Operating range of the 1,296 t/h CFB boiler during the tests

Parameter	Unit	Overall range
Superficial gas velocity	$m \cdot s^{-1}$	3.81–6.17
Terminal velocity	$m \cdot s^{-1}$	2.34–2.94
Minimum fluidization velocity	$10^{-2} \cdot m \cdot s^{-1}$	2.57–3.18
PA/SA ratio	-	1.96–4.15
Pressure drop	kPa	6.43–8.82
Bed pressure	kPa	6.0–8.2
Furnace temperature	°C	760–879
Bed temperature	°C	769–863

Table 3: Coal characteristic for all test runs

Parameter	Unit	Overall range
Proximate analysis (as received)		
LHV	MJ/kg	21.73–23.87
Ash	%wt.	9.70–12.87
Moisture	%wt.	11.93–14.37
Volatile matter	%wt.	29.40–30.47
Ultimate analysis (air dried basis)		
Carbon	%wt.	61.30–63.76
Hydrogen	%wt.	3.20–3.55
Nitrogen	%wt.	1.01–1.04
Oxygen	%wt.	5.99–6.42
Sulphur	%wt.	1.12–1.42

INTREX™ chamber, feeding points for fuel and sorbent, make-up sand and recirculation fly ash.

For NO_x emission reduction, the boiler was equipped ammonia injection system. Ammonia water line is led from storage tank to the ammonia pumps, which pump the NH_3 to the upper part of the each solids separator. The ammonia injection ports are located on the sidewalls of the solids separators. For each solids separator 4 injection ports will be reserved that are located on four different levels from the grid. Pressurized air shall assist in solution atomizing and keep the spray nozzles clean during boiler operation. The ammonia water droplet size is controlled by adjusting the atomizing air.

4. Experimental conditions

Before performance tests lasting eight hours, the operating conditions of the CFB combustor were stabilized for four hours. When stable operating conditions in the CFB combustor were achieved, four measurement series were conducted. One measurement serie was generally run for ca. 2 hours. In the experiments, supercritical circulating fluidized bed boiler runs under the condition of 40% to 100% full load. During all test runs, the boiler was operated in a steady state i.e. feed rate, bed pressure, bed temperature, air flow rate, O_2 concentration within combustion chamber all being constant. All the data were collected on-line and the arithmetic average values were used in the paper. The main parameters of the data process for supercritical CFB boiler during all test runs is given in Table 2.

In the Table 2, the terminal velocity U_t was calculated as follows [23]:

$$U_t = \left[4d_p\left(\rho_s - \rho_f\right)g\right] \cdot \left[3\rho_f C_D\right]^{-1} \tag{1}$$

where C_D is the drag coefficient predicted by Eq. (2), d_p denotes the mean particle diameter and g represents the gravitational constant. The gas density is denoted by ρ_f and ρ_s represents solid density. For non-spherical particles, drag coefficient value was expressed as [23]:

$$\begin{aligned} C_D = &\left(\tfrac{24}{Re_p}\right)\left[1 + 8.172e^{(-4.066\phi)}Re_p^{0.0964+0.557\phi}\right] \\ &+ \left[\left(73.69Re_p e^{-5.075\phi}\right)\left(Re_p + 5.378e^{6.212\phi}\right)^{-1}\right] \end{aligned} \tag{2}$$

where Re_p is the particle Reynolds number and ϕ represents the spherecity. In the Table 3, the ratio between the primary air to the secondary air is denoted by PA/SA. The pressure drop Δp based on the measurement of pressure data along height of furnace chamber and was obtained by:

$$\Delta p = (p_i - p_{i+1}) \cdot (\Delta H)^{-1} \tag{3}$$

where p_i and p_{i+1} is local pressure data for bottom region and exit region of combustion chamber, respectively. Moreover, ΔH represents difference of height between two levels above grid.

The coal used in the experiments is a Polish bituminous coal and the proximate and ultimate analysis data are given in Table 3.

The particle size range of the coal samples is 0.07–13.75 mm. The mean diameter of bituminous coal is 2.64 mm.

The load, pressurized air and added amount of NH_3 were each sampled every 15 min. The deNO$_x$ system was operating at different ammonia mass flows and at three different pressurized air levels (i.e. low, mid and high). A spray of water with 25%wt. ammonia was injected in the gas from a nozzle at two levels of the sidewall separator. Ammonia water solution flow was evenly distributed to all solids separators. Ammonia consumption was scaled to the design point using linear correction from fuel nitrogen.

Design fuel has 0.83% nitrogen. Fuel used in test runs had 1.04% nitrogen. Excess nitrogen in fuel causes 25.3% higher NO_x emissions. Flue gas composition and the gas temperature inside separators were continuously measured and stored as 5 min averages using the online stack monitoring. The simple point is located in the horizon flue gas duct before the electrostatic precipitator. At the different boiler loads, the flue gas temperature within separators was typically in the range of 636° to 845°C, whereas the flue gas O_2 concentration varied between 2.88% and 7.11%. Moreover, the oxygen concentration was measured in the upper region of furnace chamber with a dilute phase of solids.

The effective efficiency of deNO$_x$ system was calculated using following Eq. (4):

$$\eta_{NOx} = 100\% \cdot (X_{NOx,\,out\,NH3} - X_{NOx,\,in\,NH3}) \cdot (X_{NOx,\,out\,NH3})^{-1} \quad (4)$$

where η_{NOx} is denoted total efficiency of deNO$_x$ system [%], $X_{NOx,\,out\,NH3}$ represents inlet concentration of NO_x without ammonia injection to flue gas [mg/m$_n^3$] and $X_{NOx,\,in\,NH3}$ means outlet concentration of NO_x from deNO$_x$ system [mg/m$_n^3$].

5. Results and discusion

The effect of boiler load on NO_x emission is presented in Fig. 2.

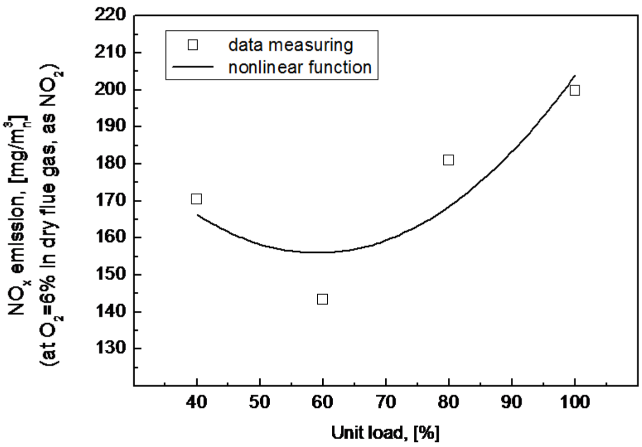

Figure 2: Nitric oxides levels for different unit loads

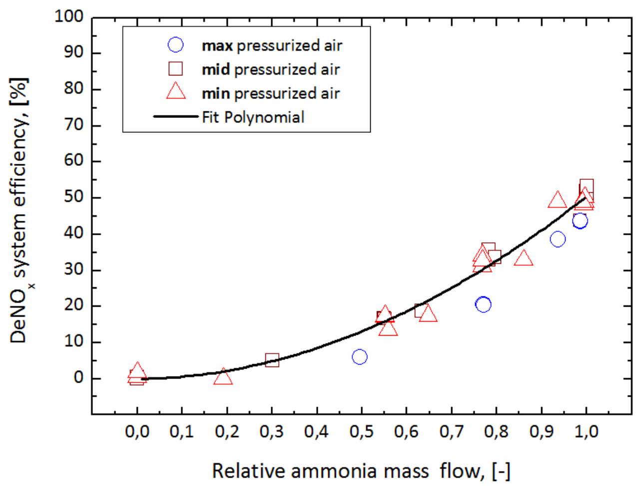

Figure 3: Effect of ammonia mass flow on deNO$_x$ system efficiency

Low NO_x concentrations were obtained at 60% MCR unit load because optimum flue gas temperature occurs inside separators to reduction of NO_x by means of ammonia. The results confirmed that under operational conditions more attention should be focused on the effect of the flue gas temperature in the "thermal deNO$_x$" process. Flue gas temperature inside solids separator and temperature in upper part of the furnace chamber were dependent on boiler load. At higher load was recommended injection of bigger amount of ammonia for upper injection ports, and during low load operation ammonia injection to lower injection ports.

The results of deNO$_x$ system efficiency against NH_3 mass flow for varying presurized air spray at

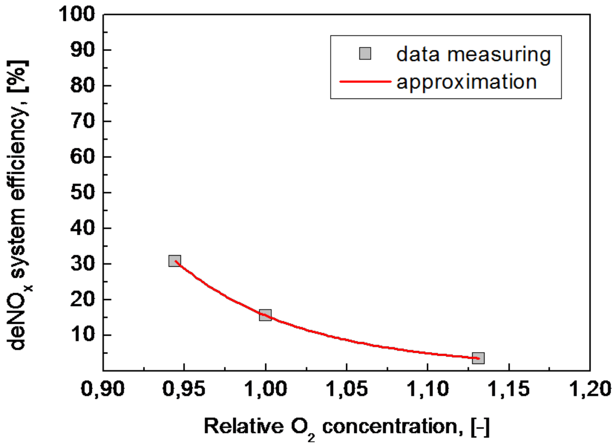

Figure 4: Efficiency of deNO$_x$ system for different oxygen concentration in the furnace chamber at the full load boiler

100%MCR unit load are plotted in Fig. 3. Ammonia injection tests were carried out at the steady mass flow for granular materials supply to boiler and constant bed pressure.

From the Fig. 3 it can be observed that ammonia mass flow remarkable affects the NOx emission. The relative ammonia mass flow was calculated using following Eq. (5):

$$X_{NH3} = \frac{X_{NH3\,act}}{X_{NH3\,ref}} \quad (5)$$

where X_{NH3} is denoted relative ammonia mass flow and $X_{NH3\,act}$ represents measured and $X_{NH3\,ref}$ reference ammonia mass flow to flue gas.
An increase in relative ammonia mass flow from 0.22 to 1.00 results in an increase in NO$_x$ removal from flue gas (ca. 53%). Results show in Fig. 3 that a reduction of NO$_x$ emission was possible at a NH_3/NO$_x$ molar ratio of 1.52–6.18. A spray pressure of air with 25%wt. ammonia not affects on NO$_x$ emission. This is because nearly the constant reaction time between NO$_x$ and NH_3 and uniform distribution of injected ammonia. During all test runs it was possible to achieve permissible nitric oxides levels according to environmental regulations for standard emission (i.e. 200 mg/m$_n^3$) when fuel specific amount of ammonia injection was used.

The effect of oxygen concentration within combustion chamber on deNO$_x$ system efficiency is shown in Fig. 4. In this paper, three relative values of O_2 concentration (i.e. 0.94, 1.0 and 1.13), were studied at the same amount of the ammonia injection to flue gas. The relative oxygen concentration was calculated using following Eq. (6):

$$X_{O2} = \frac{X_{O2\,act}}{X_{O2\,ref}} \quad (6)$$

where X_{O2} means relative O_2 concentration in the flue gas and $X_{O2\,act}$ is measured and $X_{O2\,ref}$ is reference O_2 concentration in the flue gas, %.
As oxygen concentration in the flue gas increase, the NO$_x$ emission gradually increases and had nonlinear uptrend. As shown in Fig. 4, by operating the boiler with the nominal flue gas O_2 concentration strict NO$_x$ emission requirements can be fulfilled. The highest O_2 concentration promoted conversion of the fuel-N to NO$_x$. In the SNCR process, the ammonia oxidation rate depends on the O_2 in the ammonia injection zone. The thermal NO$_x$ reduction rates is sensitive to oxygen concentration in the flue gas. When ammonia was injected into the downstream flue gas, NO was converted into harmless molecular nitrogen and water vapor by the following homogeneous reaction:

$$4NH_3 + 6NO \rightarrow 5N_2 + 6H_2O \quad (7)$$

Moreover, a low-oxygen level in the flue gas, resulting in lower conversion of fuel-N to nitrogen oxides and strongly depends on the excess oxygen concentration. During combustion at low oxygen concentration, low char content in the bed and CO concentration (i.e. 53 mg/Nm^3) leading to reduction of NO by the following reactions:

$$2NO + 2C \rightarrow N_2 + 2CO \quad (8)$$

$$2NO + 2CO \rightarrow N_2 + 2CO_2 \quad (9)$$

The Figure 5 illustrates the influences of flue gas temperature on efficiency of deNO$_x$ system at NH_3/NO$_x$ molar ratio 3.85.
The experimental results indicate the optimum temperature window (i.e. from 720°C to 790°C) for NO$_x$ reduction by means of NH_3. Whereas the flue gas temperature was too high, ammonia oxidized to NO$_x$ and ultimately to reduced the efficiency of SNCR system. If the flue gas temperature was too low, the NO$_x$ removal process was diminished and unreacted ammonia concentration increased in exhaust

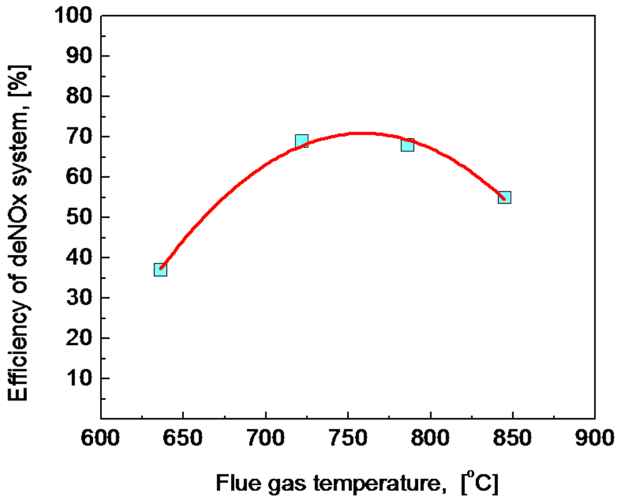

Figure 5: $DeNO_x$ system efficiency as a function of flue gas temperature

gases. The highest measured of NO_x abatement was the nearly 70%. In the case of 40% boiler load, NO_x reduction was the lowest (i.e. 37%) because the flue gas temperature was too low for sufficient reaction between NH_3 and NO_x.

6. Conclusions

The effects of operation parameters on NO_x emission in supercritical CFB boiler are quite important but rarely reported. In the paper, parameters including ammonia mass flow, a spray pressure of air, O_2 concentration and flue gas temperature were studied in a 1296 t/h Lagisza CFB boiler. The field experiment shows that the ammonia mass flow significantly affects on $deNO_x$ system efficiency. The permissible nitric oxides levels was easily achieved with fuel specific amount of ammonia injection. Results show that $deNO_x$ system efficiency does not depend upon a spray pressure of air with 25 %wt. ammonia water. The oxygen concentration significantly affects on $deNO_x$ system efficiency. Elevating O_2 concentration about 20% within furnace chamber cased $deNO_x$ system efficiency about 25% lower. Large-scale measurements confirm the existence of a maximum the efficiency of NO_x removal around 70% when using ammonia at flue gas temperature in the range from 720°C to 790°C. $DeNO_x$ system provides an efficient way to reduce NO_x emission in supercritical circulating fluidized bed boiler.

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