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PILOT-SCALE STUDIES ON NO_X REMOVAL FROM FLUE GAS VIA NO OZONATION AND ABSORPTION INTO NaOH SOLUTION

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The paper presents results of experimental studies on removal of NO_x from flue gas via NO ozonation and wet scrubbing of products of NO oxidation in NaOH solutions. The experiment was conducted in a pilot plant installation supplied with flue gas from a coal-fired boiler at the flow rate 200 m^3/h. The initial mole fraction of $NO_{x,ref}$ in flue gas was approx. 220 ppm, the molar ratio $X = O_3/NO_{ref}$ varied between 0 and 2.5. Ozone (O_3 content 1÷5% in oxygen) was injected into the flue gas channel before the wet scrubber. The effect of the mole ratio X, the NaOH concentration in the absorbent, the liquid-to-gas ratio (L/G) and the initial NO_x concentration on the efficiency of NO_x removal was examined. Two domains of the molar ratio X were distinguished in which denitrification was governed by different mechanisms: for $X \leq 1.0$ oxidation of NO to NO_2 predominates with slow absorption of NO_2, for $X >> 1.0$ NO_2 undergoes further oxidation to higher oxides being efficiently absorbed in the scrubber. At the stoichiometric conditions ($X = 1$) the effectiveness of NO oxidation was better than 90%. However, the effectiveness of NO_x removal reached only 25%. When ozonation was intensified ($X \geq 2.25$) about 95% of NO_x was removed from flue gas. The concentration of sodium hydroxide in the aqueous solution and the liquid-to-gas ratio in the absorber had little effect on the effectiveness of NO_x removal for $X > 2$.

Keywords: de-NOx, nitric oxide, ozonation, absorption

1. INTRODUCTION

The most abundant gaseous air pollutants emitted from coal-fired power plants are sulphur dioxide (SO_2) and nitrogen oxides (NO_x) (Air quality in Europe, 2011). For the reduction of NO_x emission from coal-fired boilers combustion modification systems such as: low-NO_x burners (LNB) and high temperature air combustion (HTAC) (Budzianowski and Miller, 2009), reburning (Smoot et al., 1998; Werle, 2012), over fire air (OFA) and rotating opposed fired air (ROFA) (Błasiak, 2010) have been developed and are commonly used in coal-fired power plants (Spalding et al., 2006). New strict demands, applied by the EU, concerning NO_x emission values made it necessary to apply more efficient post-combustion methods of flue gas denitrification (Directive2010/75EU, 2010).

In the developed EU countries the emission values for NO_x from coal-fired power plants are controlled applying the selective catalytic reduction (SCR). The SCR is a very effective method of NO_x emission control and has got a status of the Best Available Technology (BAT). However, this method has also its weak points. Its capital and exploitation costs are considerably high (Krotla et al., 2009) moreover, the SCR installation can cause problems in the maintenance of pulverised coal-fired boilers. The temperature of de-NO_x process in SCR is high (430÷470 ºC), and therefore requires additional flue gas heat exchangers. The live-time of the catalysts is limited due to intensive fly ash erosion (Van der Kooij

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et al., 1997). When biomass is co-fired catalysts could be poisoned by alkali metals. Moreover, the use of ammonia in SCR may induce risk of the ammonia-slip.

Promising alternatives to the SCR are processes for simultaneous removal of NO_x and Hg, which could be combined with wet methods of flue gas desulfurization (FGD) (Ellison, 2003). These methods are based on preliminary low-temperature oxidation of weakly soluble NO and absorption of higher nitrogen oxides in alkaline solutions. During the last two decades several oxidizers were examined regarding their capability of NO oxidation, safety and economic issues. Chironna and Altshuler (1999) discussed the chemical aspects of NO_x scrubbing considering oxidants: O_2, O_3, ClO_2 and NaOCl. Nelo et al. (1997) studied the simultaneous oxidation of NO_x and SO_2 by ozone and hydrogen peroxide and noticed that ozone practically does not oxidize SO_2 at the room temperature. Gostomczyk and Krzyżyńska (2005) examined the effectiveness of simultaneous removal of NO_x, SO_2 and Hg from flue gas using gaseous (O_3) and aqueous (NaOCl and H_2O_2) oxidants. Chen et al. (2005) studied oxidation and absorption of NO applying sodium hypochlorite aqueous solution in a packed tower. Hutson et al. (2008) conducted bench-scale study on simultaneous removal of NO_x, SO_2 and Hg by an addition of sodium chlorite ($NaClO_2$) into a wet $CaCO_3$ scrubber.

Among the considered oxidizers ozone appeared to have many advantages and most of work has been done on its use for the NO_x emissions control. The process of NO oxidation by ozone in the well-stirred reactor was numerically studied by Puri (1995). Nelo et al. (1997) showed that for efficient removal of NO_x substantial ozone excess is required. Chironna and Altshuler (1999) suggested that slow oxidation rate of nitrogen oxide by air could be greatly improved by adding ozone. Jaroszynska-Wolińska (2002) showed a significant acceleration of NO removal from waste gases in a two-stage oxidation-absorption process by ozone addition. Cannon Technology Inc. in collaboration with BOC Gases developed a low temperature oxidation (LTO) for NO_x removal by ozone injection (Jarvis et al, 2003). Fu and Diwekar (2003) conducted the cost-effectiveness analysis of the LTO process. Mok (2006) and Mok and Lee (2006) examined experimentally a two-stage ozonation-wet reduction process of NO_x removal in which NO_2 was reduced by sodium sulphide. More than 95% of removal efficiency was achieved. Wang et al. (2007) performed lab-scale studies on the oxidation-absorption process of NO, SO_2 and Hg^0 applying ozone. They proved the possibility of simultaneous capturing of NO_x and SO_2 as well as 80% oxidations of elemental mercury. Sun et al. (2011) studied the process of NO oxidation by ozone and absorption of NO_2 and SO_2 with pyrolusite slurry (MnO_2 ore) in a bubbling reactor. Jaroszyńska-Wolińska (2009) studied numerically the chemical mechanism of the nitrogen oxide oxidation by ozone. Skalska et al. (2011a) made direct measurements of the NO ozonation products. Skalska et al. (2011b) proposed a kinetic model of NO ozonation and the rate constants based on the lab-scale experiment. The effect of ozone on exhaust emissions from combustion processes was also studied (Wilk and Słupek, 2005).

However, commercialisation of the method has met some economic obstacles, mainly because ozone generation is expensive due to oxygen demand and high energy-consumption. Further studies are necessary in order to reduce the costs of ozonation by optimisation of the ozone use.

This paper is one of a few describing pilot plant scale studies into NO_x removal from flue gas via NO ozonation and absorption of higher nitrogen oxides. Most attention was devoted to the influence of the molar ratio O_3/NO_{ref}, the initial concentration of NO_{ref} and the absorption conditions on the efficiency of NO_x removal. Additionally, the observed discrepancy between de-NO_x effectiveness attained in the lab- and pilot-scale was considered.

2. CHEMICAL KINETICS OF NO OZONATION BY OZONE

Nitrogen oxide, which is the main component of NO_x, is relatively nonreactive. In the atmosphere it is oxidized by oxygen and ozone to more reactive nitrogen dioxide NO_2, which is next converted into nitric acid and nitrites removed from the atmosphere with acid rains (Prather and Logan, 1994). Knowledge about the atmospheric chemistry appeared to be helpful for developing the low-temperature method of NO_x abatement (Anonymous, 2001).

The reduced set of chemical equations used in the study in order to explain the governing mechanisms of NO ozonation and interpret the experimental results is presented in Table 1.

Table 1. Reaction rate constants (Skalska et al., 2011b)

Reactions	Value of k_f/k_b (298 K), dm^3, mole, s	No.
$NO + O_3 \rightarrow NO_2 + O_2$	$1.08 \cdot 10^7$	(1)
$NO + NO_2 = N_2O_3$	$4.76 \cdot 10^9$ / $3.6 \cdot 10^8$ (NIST, 2012)	(2)
$NO_2 + O_3 \rightarrow NO_3 + O_2$	$2.39 \pm 0.14 \cdot 10^4$	(3)
$NO_2 + NO_3 = N_2O_5$	$3.16 \pm 0.61 \cdot 10^4$ / $3.51 \pm 0.71 \cdot 10^{-3}$	(4)
$N_2O_5 + H_2O \rightarrow 2HNO_3$	$2.43 \pm 0.34 \cdot 10^{-3}[H_2O]^{-1}$	(5)
$2O_3 + M \rightarrow 3O_2$	depends on specific M	(6)

The reaction of NO oxidation (1) is very fast. The forward and backward reactions (2) are very fast as well, but the reaction product (N_2O_3) is unstable, and therefore ignored in most modelling studies (Wang et al., 2006; Jaroszyńska-Wolińska, 2009). However, N_2O_3 may play an important role in the absorption process (Głowiński et al., 2009).

When the molar ratio X of ozone and the reference nitrogen oxide (X = O_3/NO_{ref}) reaches the sub-stoichiometric values ($X < 1$) nitrogen dioxide is the main product of NO oxidation (Nelo et al., 1997). When the ozone mole fraction grows to the over-stoichiometric values ($X > 1$) the reaction (3) of NO_2 and overdosed O_3 becomes important because of NO_3 radicals formation. For more intensive NO ozonation (X >> 1) nitrogen trioxide reacts with NO_2 to form dinitrogen pentoxide N_2O_5 (Skalska et al., 2011b).

The reaction (6) was included into the scheme (Table 1) in order to emphasise an increase of ozone demand due to ozone losses in aside reactions, including those with carbon monoxide, steam, dust particles and channel walls in industrial applications.

It is generally accepted that NO has a very low solubility (Nelo et al., 1997; Skalska et al., 2011a; Wang at al., 2007). Although nitrogen dioxide has better solubility than NO, it is still not sufficient for effective removal in wet scrubbers (Joshi et al., 1985). Moreover, the absorbed NO_2 reacts with water producing nitrous and nitric acid:

$$2NO_{2(l)} + H_2O \rightarrow HNO_2 + HNO_3 \qquad (7)$$

but nitrous acid HNO_2 is unstable in the presence of strong acids such as HNO_3 and can undergo decomposition releasing NO (Thielmann et al., 2005):

$$3HNO_2 \rightarrow HNO_3 + 2NO + H_2O \qquad (8)$$

These are the possible reasons that for sub-stoichiometric ozonation ($X \leq 1$) the effectiveness of NO_x removal is limited to approx. 20% (Jakubiak and Kordylewski, 2010; Nelo et al., 1997). More intensive ozonation (X >> 1) leads to formation of dinitrogen pentoxide, which is highly water-soluble and its reaction with water (5) gives stable nitric acid HNO_3 (Bertram and Thornton, 2009).

When conversion of NO into N_2O_5 is required, the stoichiometric ozone demand results from the summary chemical reaction:

$$NO + 3/2O_3 = 1/2N_2O_5 + 3/2O_2 \tag{9}$$

It means that the stoichiometric molar ratio O_3/NO_{ref} should be X = 1.5 which is 50% larger than that based on Eq. (1).

3. EXPERIMENTAL

The pilot plant installation used in the experiment was supplied with flue gas by the fan 15 (200 m^3/h) from the pulverised coal-fired boiler OP-430 (Fig. 1). The installation, designed for general purpose investigations on flue gas cleaning, was used only in a limited range in these studies. Ozone was injected into the flue gas duct between the fabric filter 4 and the absorption column 19. The absorber 19 was a column of the inner diameter d_i = 190 mm and the height 4 m with the container of sorbent 12 below. The absorbent was an aqueous solution of sodium hydrate (NaOH), which was injected into the absorber column through nozzles 11, 17, 18 and 20 under the pressure of 0.2 MPa on four levels.

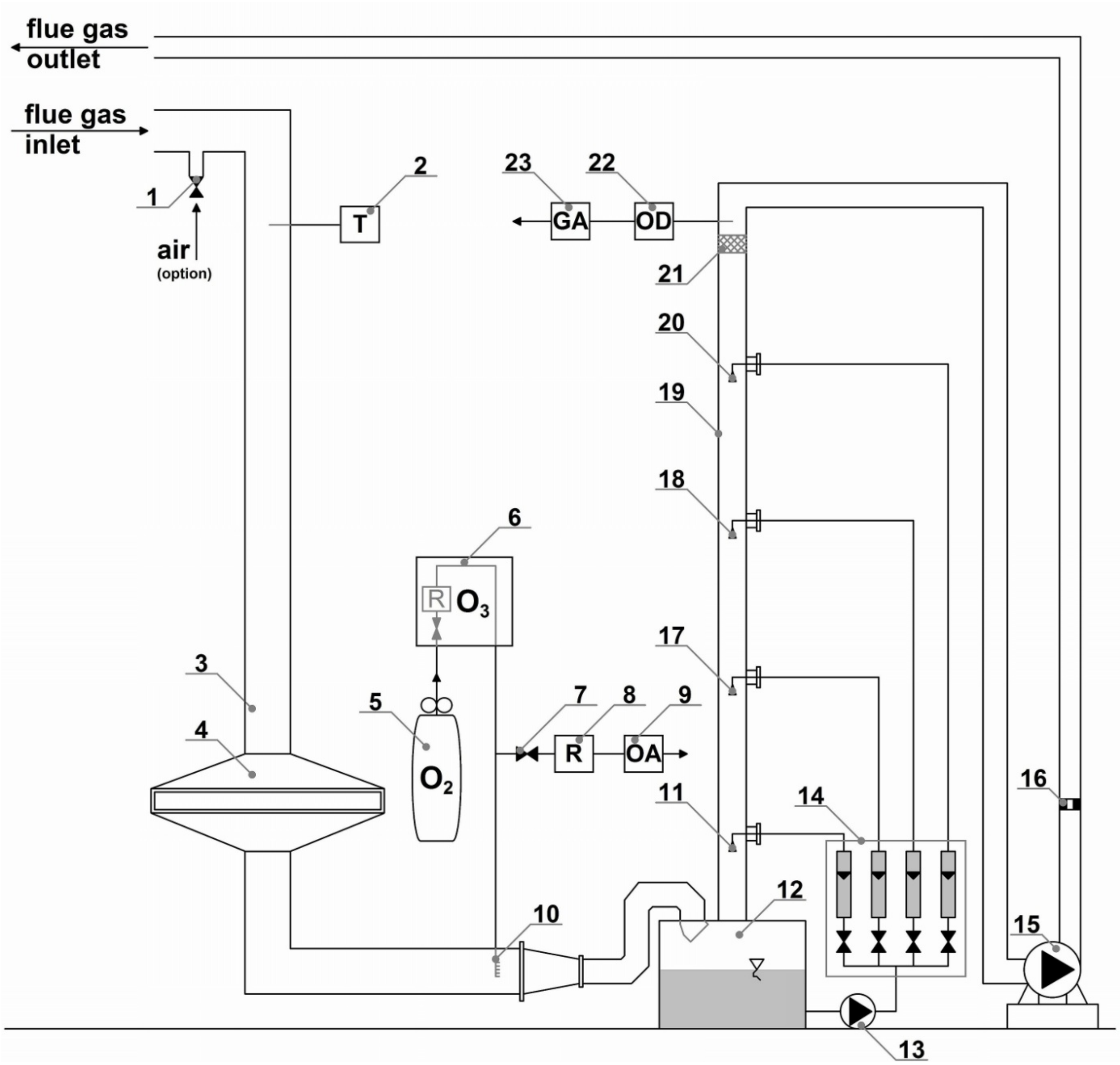

Fig. 1. Scheme of the pilot plant;
1, 7 – valves, 2 – electronic thermometer (PT-100), 3 – flue gas duct, 4 – fabric filter, 5 – steel cylinder of O_2, 6 – ozone generator, 8, 14 – rotameters, 9 – ozone analyser, 10 – ozone lance, 11, 17, 18, 20 – nozzles, 12 – container, 13 – pump, 15 – fan, 16 – measuring orifice plate, 19 – absorption column, 21 – demister, 22 – ozone destructor, 23 – gas analyser

Ozone (1÷5% O_3 in oxygen by volume, depending on the required molar ratio X) was injected into flue gas under the pressure of 0.07 MPa by a lance 10 through five nozzles at the flow rate of 2.6 m^3/h. The oxidising reactor was approximately horizontal duct connected to the container of sorbent 12. The residence time in the oxidising reactor was approx. 2 s. The inflow temperature of flue gas (about 90 ºC) was reduced by a gas cooler to maintain the temperature of approx. 40 °C which is required before CO_2 capture installation. The oxidation and absorption processes were conducted at the temperature of approx. 40 and 35 °C respectively.

Ozone was produced by the ozone generator 6 of the type OZAT CFS-3 2G of Degremont Technologies Ltd (Ozonia) which was fed by oxygen (2.6 m^3/h) from the steel cylinder 5. The ozone flow rate into the flue gas duct was controlled by the method described elsewhere (Jakubiak and Kordylewski, 2011).

The molar fractions of NO and NO_2 in flue gas were measured after the absorber demister 21 by the gas analyser 23 Testo 350xl of Testo Inc. The reference molar fractions of NO_{ref} and $NO_{x,ref}$ denoted the molar fractions of NO and NO_x measured in flue gas after the absorber when ozone was not generated in oxygen flowing through the ozoniser 6.

A series of experiments were conducted in order to examine the influence of the molar ratio O_3 to NO_{ref} (X), the concentration of NaOH in the scrubbing solution, liquid-to-gas ratio (L/G) in the absorber and the initial concentration of NO in flue gas on the effectiveness of NO oxidation (OR) and NO_x removal (η). The basic parameters related to the conditions of the performed experiments are presented in the Table 2.

Table 2. Selected parameters used in the experiments

Parameter	Unit	Value
Volumetric flow rate of flue gas	m^3/h	200
Volume concentration of NO in flue gas	ppm	~220
Volume concentration of NO_2 in flue gas	ppm	8÷10
Volume concentration of O_2 in flue gas	%	9.5
Volumetric flow rate of O_2+O_3 mixture from the ozone generator to the ozone analyser	dm^3/h	16
Type of absorbent	-	NaOH aqueous solution
NaOH concentration in the absorbent solution	M	0÷1
Volumetric flow rate of a solution in the absorption column	dm^3/h	500÷2000
Liquid-to-gas ratio (L/G)	dm^3/m^3	2.5÷10
Flue gas temperature (inlet)	ºC	~95
Flue gas temperature in the oxidizing reactor	ºC	40
Flue gas temperature in the absorption column	ºC	35

The effectiveness of NO oxidation was determined based on the mole fractions of NO measured in flue gas after the demister 21 by following expression denoted further the oxidation ratio OR (%):

$$OR = \left(1 - \frac{[NO_{out}]}{[NO_{ref}]}\right) \cdot 100\% \tag{10}$$

The effectiveness of NO_x removal η from flue gas was defined by the following formula:

$$\eta = \left(1 - \frac{[NO_{x,out}]}{[NO_{x,ref}]}\right) \cdot 100\% \tag{11}$$

4. RESULTS

4.1. Dynamics of the NO ozonation process

Dynamics of NO ozonation and absorption were studied varying the feeding rate of ozone. Figure 2 shows the recorded response of NO and NO_2 mole fractions measured behind the demister 21 after sudden ozone supply to flue gas at the molar ratio of $X = 2.0$.

After approx. 10 s. delay the mole fraction of NO was decreasing for about 1.5 min. and finally reached the level below 10 ppm. This transition period could have resulted from a long residence time in the volume over the surface of sorbent in the container 12 (Fig. 1). At the same time the nitrogen dioxide mole fraction first increased and achieved the maximum (100 ppm) and next declined to approx. 10 ppm.

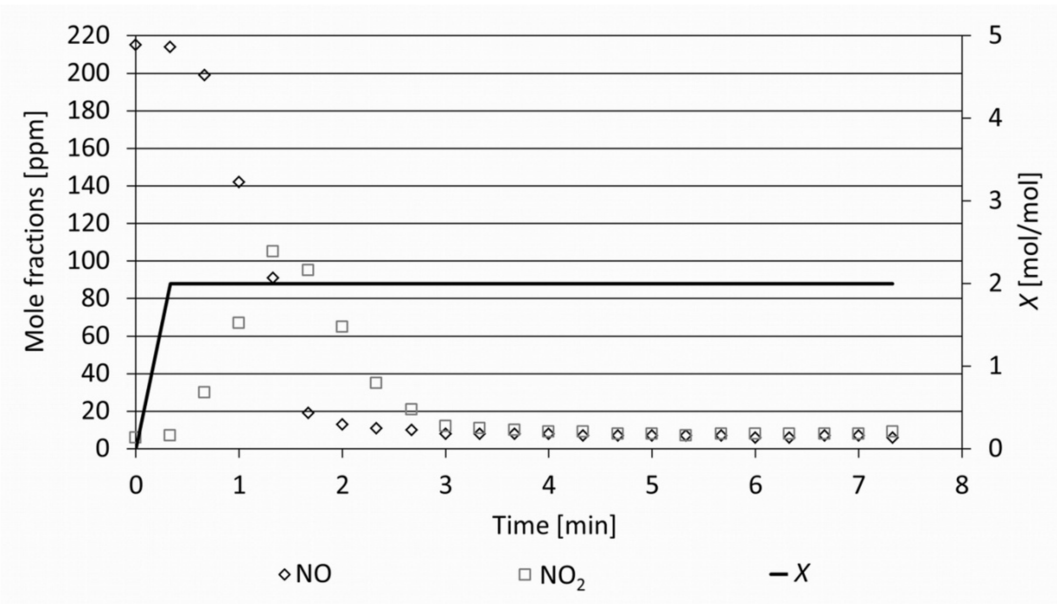

Fig. 2. Variation in time of NO and NO_2 mole fractions after the start of ozone feeding ($X = 2.0$, $L/G = 10$ dm^3/m^3, 0.1M solution of NaOH)

Time-dependent changes of NO and NO_2 mole fractions in flue gas after the absorber when the mole ratio X was gradually increasing from 0 to 2.25 are shown in Fig. 3. The nitrogen oxide mole fraction dropped almost proportionally to the molar ratio X increments up to $X \cong 1.0$. As such the oxidation rate of the residual NO (approx. 10 ppm) slowed down, perhaps because of competition from a much higher mole fraction of NO_2.

According to the chemical equation (1) the nitrogen dioxide mole fraction quickly increased achieving the maximum in the range of $X = 1.0 \div 1.25$. It was further gradually declining, and finally approached almost zero above $X \cong 2.0$. The observed difference in the behaviour of the NO_2 mole fraction can be explained on the basis of the kinetic scheme (Table 1): at the sub-stoichiometric conditions NO_2 was the dominating product of ozonation, whereas at the over-stoichiometric conditions NO_2 underwent further oxidation.

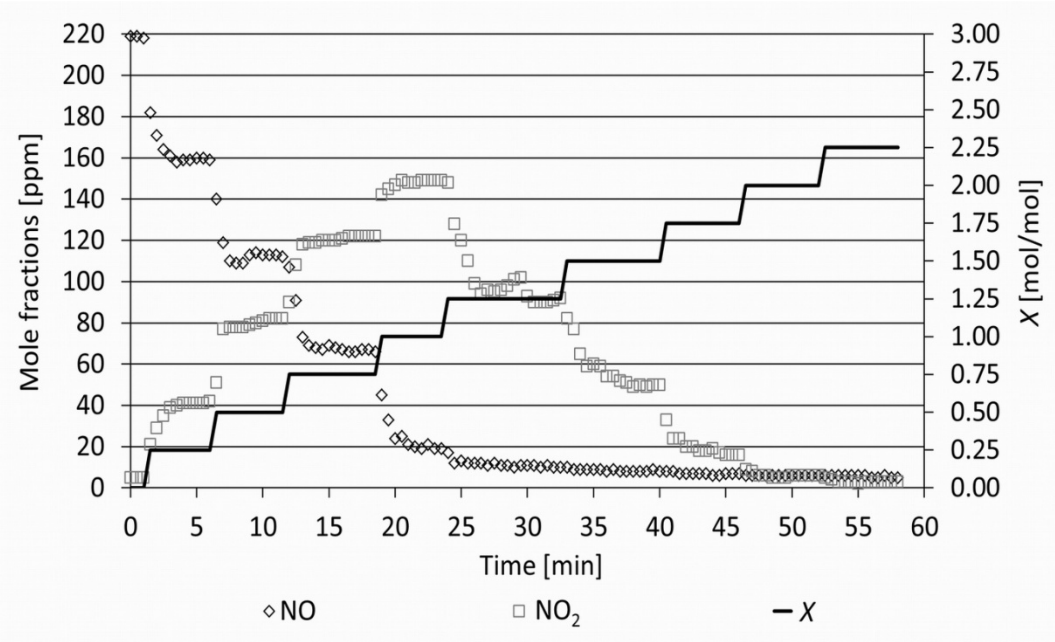

Fig. 3. Variation in time of NO and NO_2 mole fractions against the molar ratio ($X = 0 \div 2.25$, $L/G = 10$ dm^3/m^3, 0.1M solution of NaOH)

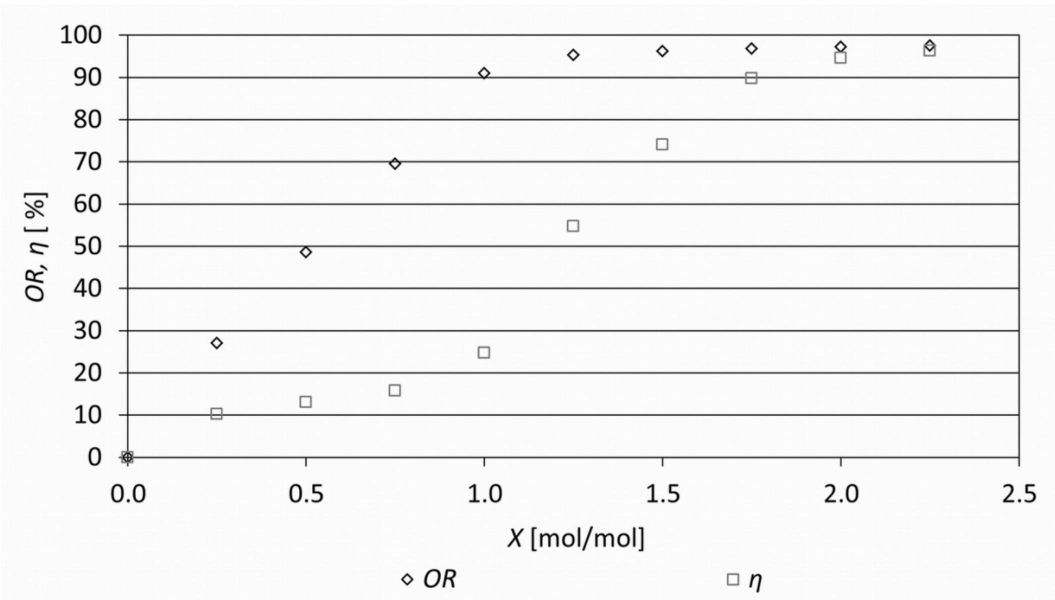

Fig. 4. Oxidation ratio *OR* and effectiveness of NO_x removal η vs. molar ratio X ($[NO_{ref}] = 219$ ppm, $L/G = 10$ dm^3/m^3, 0.1M solution of NaOH)

4.2. The effectiveness of NO_x removal vs. the molar ratio O_3/NO_{ref}

The molar ratio value X necessary to secure the needed effectiveness of NO_x removal η is an important parameter influencing the economy of flue gas denitrification. It is usually far from the stoichiometric ratio value of the NO oxidation because of some ozone losses induced by physical (mixing pattern of O_3 and NO_x and the residence time) and chemical (aside reactions) factors. The meaning of particularly the physical factors is not yet properly understood and perhaps their unfortunate choice can result in a substantial increase of the ozone excess.

The oxidation ratio *OR* and the effectiveness of NO_x removal η were calculated from the formulas (10) and (11) applying the initial (reference) and measured mole fractions of NO and NO_2. They behaved differently with the molar ratio *X* rise: the oxidation ratio *OR* was increasing almost proportionally to *X* for the under-stoichiometric values (< 1) and for $X > 1$ practically reached plateau (Fig. 4).

The effectiveness of NO_x removal η was low (< 20%) at under-stoichiometric conditions ($X \leq 1.0$). Only when the ozone flow rate increased to over-stoichiometric values ($X >> 1$) the rate of NO_x removal accelerated and achieved 90% for $X \geq 1.75$.

4.3. The absorption of NO ozonation products

The influence of NaOH concentration in aqueous solutions

Fig. 5 shows that the oxidation ratio *OR* was not sensitive to the concentration of sodium hydroxide in the absorbent at molar ratio $0 < X < 2.5$. Moreover, the effectiveness of NO_x removal η was practically not influenced by the NaOH concentration in the solution at the studied molar ratio *X*. Even water appeared to be an efficient absorbent (Fig. 6).

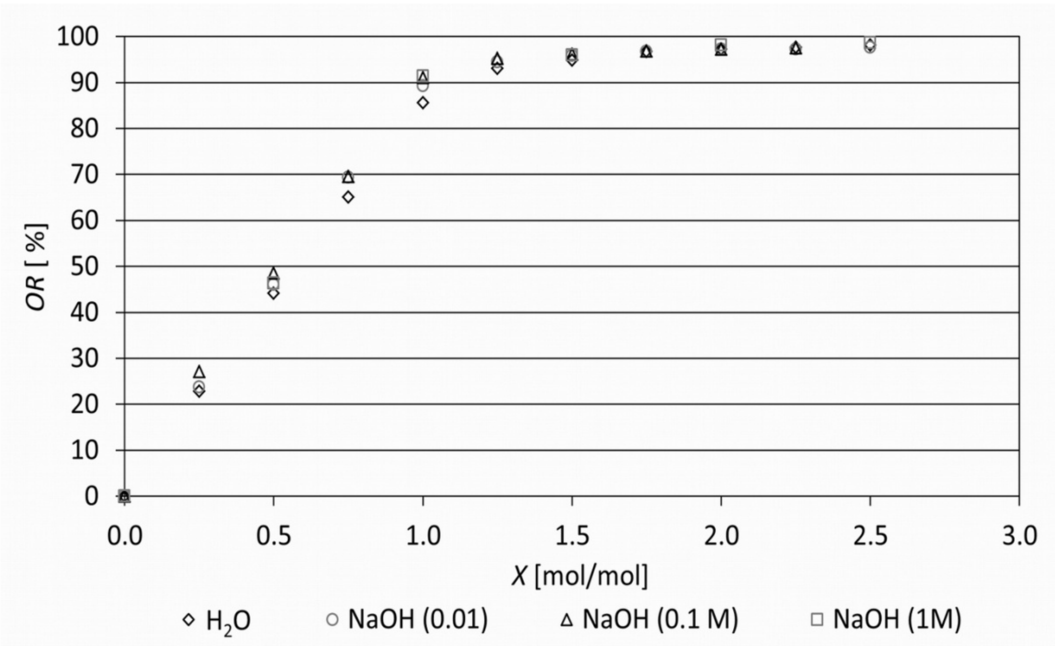

Fig. 5. Oxidation ratio *OR* vs. the molar ratio *X* depending on the NaOH concentration ($[NO_{ref}]$ = 215÷220 ppm, L/G=10 dm^3/m^3)

The impact of the liquid-to-gas ratio (L/G) in the scrubber

The influence of the intensity of sorbent spraying in the absorption column 19 on the effectiveness of NO_x removal η was examined for the two values of the molar ratio: X = 1.0 and 2.0. For the stoichiometric molar ratio (X = 1.0) only a slight increase of the effectiveness η with the L/G ratio was observed, whereas for over-stoichiometric X (2.0) the effect was practically imperceptible (Fig. 7).

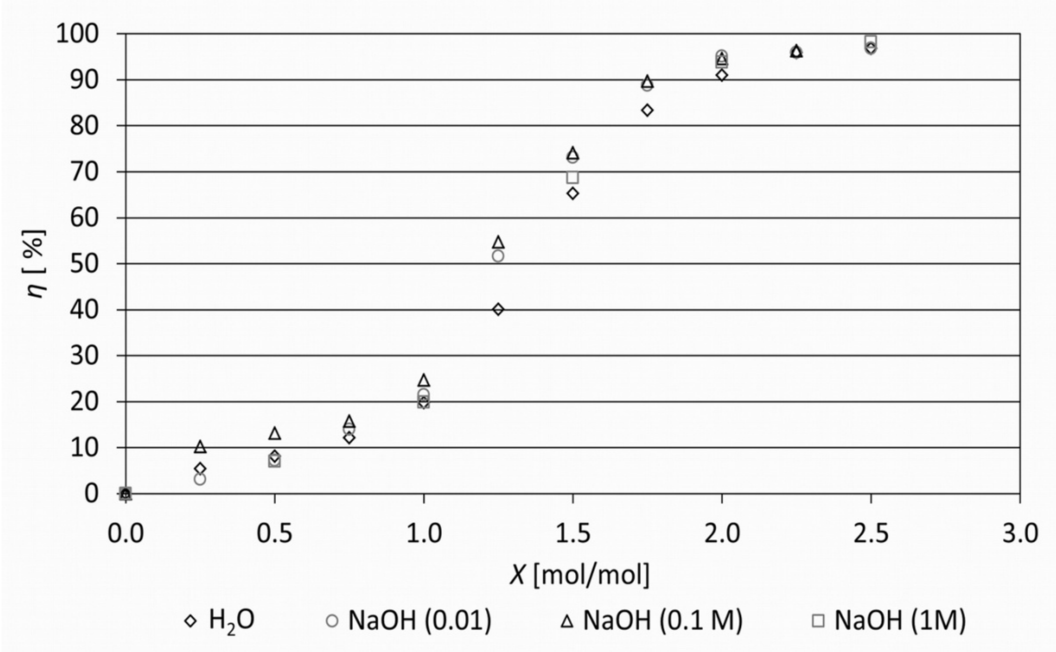

Fig. 6. Effectiveness of NO_x removal η vs. the molar ratio X depending on the NaOH concentration ($[NO_{ref}]$ = 215÷220 ppm, L/G=10 dm^3/m^3)

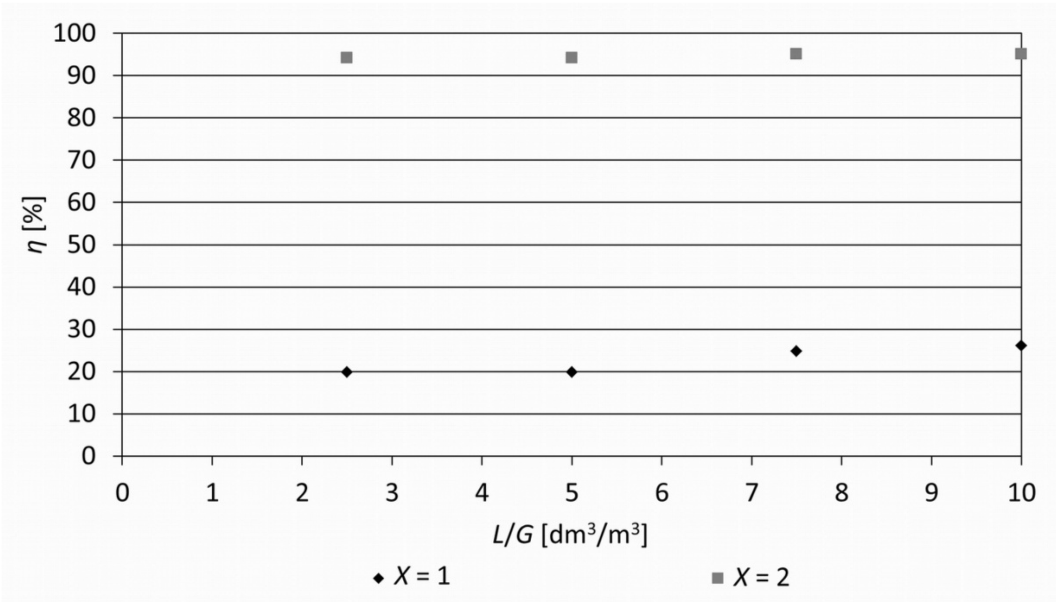

Fig. 7. Effectiveness of NO_x removal η vs. the L/G ratio depending on the molar ratio X ($[NO_{ref}]$ = 215 ppm, 0.1M solution of NaOH)

The noticed difference could be explained on the basis of different chemical mechanisms of NO oxidation in the sub- and over-stoichiometric conditions. For X = 1 the main product of NO oxidation is NO_2 which belongs to NO_x. Hence, NO_x does not change until NO_2 is absorbed in the scrubber. For X = 2.0 the dominating product of NO ozonization is N_2O_5 which does not belong to NO_x. Hence, for excessive NO ozonation NO_x is effectively removed even for low values of L/G ratio (e.g. L/G = 2.5 dm^3/m^3) (Fig. 7).

The influence of the initial NO mole fraction (NO_{ref})

The effects of the initial mole fraction of nitrogen oxide (NO_{ref}) on the oxidation ratio *OR* and the effectiveness of NO_x removal η were studied decreasing the contents of NO in flue gas by its dilution with air. The ozone feeding rate was controlled by changing the ozone mole fraction in oxygen supplied by the ozoniser 6.

Fig. 8 shows that the oxidation rate *OR* was not very sensitive to the decrease of NO_{ref} mole fraction, especially for $X = 2.0$. The slight fall in *OR* values for [NO_{ref}] approaching 50 ppm can be explained by the oxidation rate decrease for smaller mole fractions of the reactants.

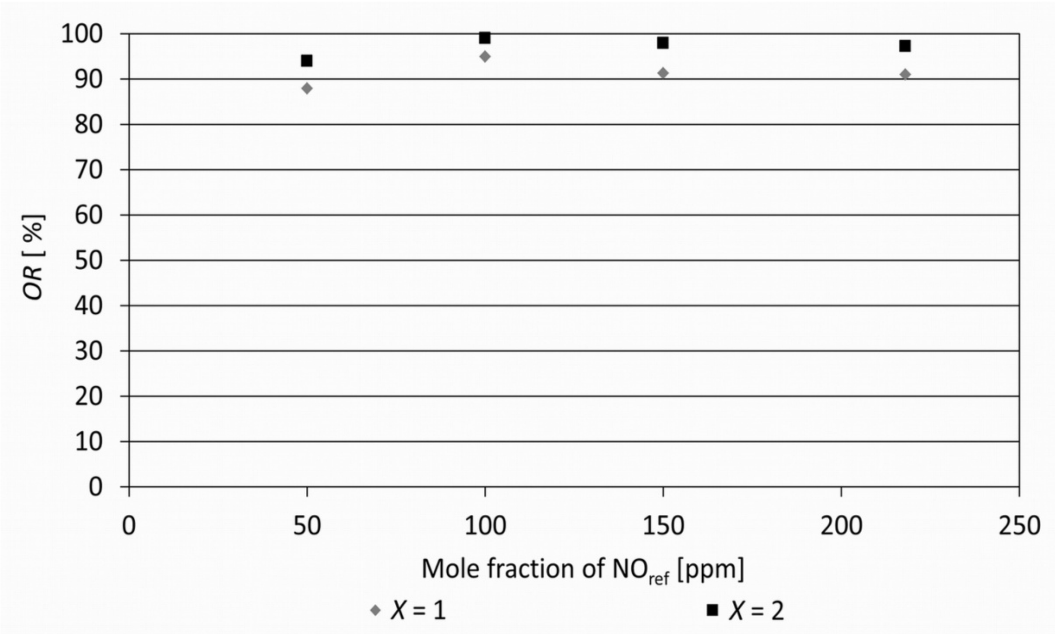

Fig. 8. Oxidation ratio *OR* vs. the reference mole fraction NO_{ref} at the molar ratios $X = 1.0$ and 2.0 ($L/G = 10\ dm^3/m^3$, 0.1M solution of NaOH)

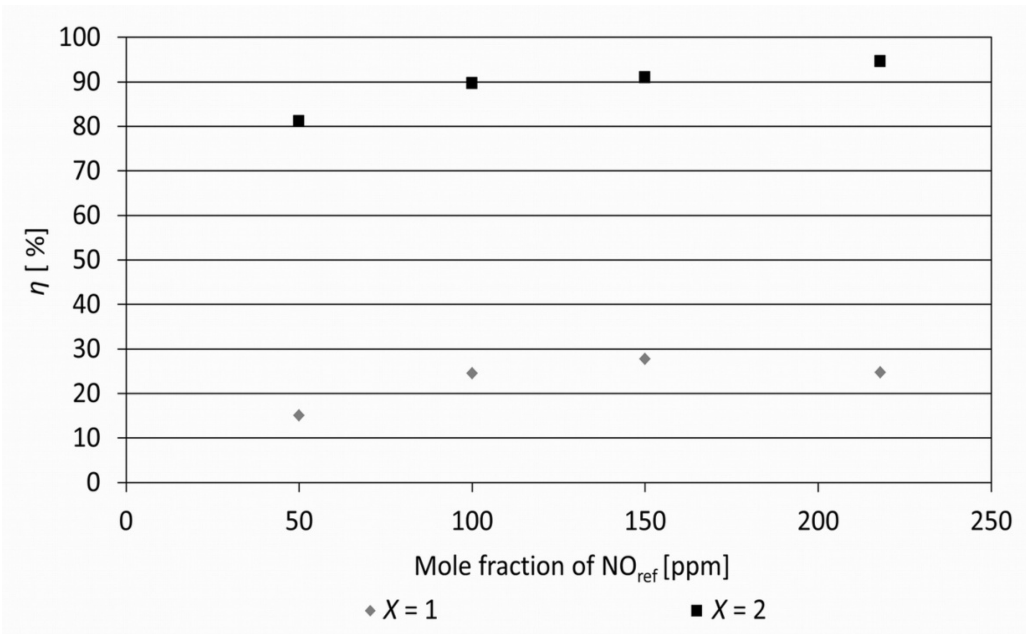

Fig. 9. Effectiveness of NO_x removal η vs. the reference molar fraction NO_{ref} for the molar ratios $X = 1.0$ and 2.0 ($L/G = 10\ dm^3/m^3$, 0.1M solution of NaOH)

The effect of the initial mole fraction NO_{ref} appeared to be more apparent for the effectiveness of NO_x removal (Fig. 9). Its more marked fall for the $[NO_{ref}]$ < 150 ppm could be accounted for by the diminished rates of the chemical reactions (1), (3) and (4) and the abated efficiency of scrubbing.

5. DISCUSSION

Having the results of the conducted studies it can be concluded that the processes of NO ozonation in flue gas can be divided into two domains depending on the molar ratio X values. For the sub-stoichiometric conditions ($X \leq 1.0$) the oxidation of NO to NO_2 was the predominating process, and its efficiency *OR* reached approx. 90%. However, the effectiveness of NO_x removal was below 20% because the absorption of NO_2 was inefficient. In the second domain of X ($X > 1.0$) the overdosed ozone also oxidized NO_2, which lead to dinitrogen pentoxide formation and the improvement of the NO_x removal above 90%.

These observations are qualitatively consistent with the results of earlier lab-scale studies where the ozonation products were absorbed in bubbling washers (Jakubiak and Kordylewski, 2010). Similar results were obtained in the lab-scale studies by other authors. Mok and Lee (2006) reported 95% efficiency of NO_x removal in their two-stage process including NO ozonation and NO_2 reduction by Na_2S. Wang et al. (2007) showed the ability to capture approximately 97% of NO_x for the molar ratio O_3/NO_{ref} = 1.6 in a system similar to that studied by Jakubiak and Kordylewski (2010). Sun et al. (2011) obtained the efficiency of NO_x removal of about 82% when applying pyrolusite slurry as an absorbent.

The scale-effect for the NO oxidation was insignificant: in the pilot-scale the oxidation ratio *OR* was approx. 90%, while in lab-scale OR = 95% for X = 1.0. This small discrepancy can be explained by more difficult conditions of ozone and NO mixing and faster consumption of ozone due to the reactions with dust, steam and carbon monoxide in flue gas.

A more distinct difference was observed for the effectiveness of NO_x removal η: in the lab-scale η it exceeded 90% for the molar ratio $X = 1.5$, whereas in the pilot-scale this value of η was achieved for $X \geq 2.0$ (Fig. 4). In this case the reason could be different absorption patterns of N_2O_5, which forms aerosol at the room temperature; perhaps it was more effectively precipitated in the bubble washer than in the scrubber.

6. CONCLUSIONS

The results of the performed experimental studies lead to the general conclusion that NO_x can be effectively removed from flue gas applying the NO ozonation and wet scrubbing. The more detailed conclusions can be formulated as follows:

- The mechanism of NO ozonation depends on the molar ratio $X = O_3/NO_{ref}$: for $X \leq 1.0$ the oxidation of NO to NO_2 is the predominating reaction, for $X > 1.0$ NO_2 undergoes further conversion and for $X \geq 1.5$ the major ozonation product is N_2O_5.
- The effectiveness of NO removal is limited by slow absorption of NO_2 for $X < 1.5$.
- The necessary condition of effective removal of NO_x by ozonation is to secure ozone excess $X > 2.0$.
- The effectiveness of NO_x removal is sensitive to the NO content; it was distinctly diminished when the initial mole fraction NO_{ref} dropped below 150 ppm.

- A change from the lab- to pilot-scale experiment resulted in an increase of the ozone demand for the same efficiency of NO_x removal.

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SYMBOLS

d	diameter of the scrubber, mm
k	reaction rate constant, dm^3, mol, s
L/G	liquid to gas ratio, dm^3/m^3
$[NO]$	NO mole fraction
$[NO_2]$	NO_2 mole fraction
$[NO_x]$	NO_x mole fraction
OR	oxidation ratio of NO, %
X	molar ratio, mol/mol

Greek symbols

η	effectiveness of NO_x removal, %

Superscripts

b	backward
f	forward
i	inner
out	output
ref	reference
l	in liquid

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