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Efficiency Increase of Secondary DeNOx Systems for Cleaning of Flue Gas Produced in Combustion Processes

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Due strict legislation providing for emission limits and increasing charges for emissions, their producers are forced to implement new technologies and methods which decrease amount of released emissions as low as possible. Nitrogen oxides (NOx) are mostly eliminated using the method of selective non-catalytic reduction (SNCR) and selective catalytic reduction (SCR). A commonly presented efficiency of NOx reduction for flue gas cleaning systems using SNCR ranges from 40 to 70 % and 90 to 94 % for cleaning systems using SCR. There are several ways of increasing efficiency of flue gas cleaning systems which mostly depend on economical financial balance. Basic methods of efficiency increase include:

- rearrangement of operating conditions of current technologies
- rearrangement of existing technological equipment
- implementation of brand new technologies totally different from original technology (most commonly replacement of SNCR system for SCR system)
- supplementing existing technology with a new one and thus increasing its efficiency

This paper presents possibility to increase efficiency of SNCR system using combination with SCR catalytic filtration or SCR fixed bed catalytic reactor to the value reaching 98 %. Possibilities of both options will be given in example for potential application at a municipal solid waste (MSW) incineration plant with waste processing capacity of 15 t/h.

1. Introduction

Combustion of various types of fuels and waste produces large amount of harmful nitrogen oxides (NOx). This can be prevented by taking primary measures. However, industrial and municipal waste is usually a heterogeneous mixture of unstable composition and these primary measures cannot be applied. Therefore, it is more proper to engage secondary measures (technologies) for already produced NOx.

NOx is usually reduced by technologies presented in Tab. 1. This comprises the so called selective non-catalytic reduction (SNCR) of NOx and selective catalytic reduction (SCR) of NOx. Selective reduction involves injection of NH_2-X compounds

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(where X = H, CN or $CONH_2$) into the stream of flue gas. NOx is then decomposed to nitrogen (N_2) and water (H_2O). Most common reaction agent is 25 % water solution of ammonia (NH_4OH) or pure ammonia (NH_3). Among other types of reaction agents there are urea water solution, nitrolime or cyanamide (European IPPC Bureau, 2003).

SNCR employs injection of reaction agent into flue gas of 930 to 980°C temperature (urea is injected into area of 950 to 1050°C temperature). Injection is usually carried out after combustion in secondary combustion chamber prior to following flue gas cleaning. Ammonia based reaction agent is dosed in stoicheiometric ratio of NH_3/NO from 0.5 to 0.9 mol/mol (exceptionally to 1.2 mol/mol) (European IPPC Bureau, 2003). Nitrogen oxides are then reduced according to stoicheiometric Equations (1) and (2) (Schnelle and Brown, 2001).

$$4NO + 4NH_3 + O_2 \rightarrow 4N_2 + 6H_2O \qquad (1)$$

$$2NO_2 + 4NH_3 + O_2 \rightarrow 3N_2 + 6H_2O \qquad (2)$$

SCR leads gas and injected agent through catalyst in temperatures from 175 to 600°C (Heck et al., 2002) depending on type and category of the catalyst. Catalyst may be a fixed bed or part of filtration bags (e.g. Remedia® fabric filtration, Cerafil TopKat® ceramic filtration). Catalysts include noble metals such as $Pt/\chi\text{-}Al_2O_3$, metal oxides such as V_2O_5/TiO_2, $V_2O_5\text{-}WO_3/TiO_2$, $V_2O_5\text{-}MoO_3/TiO_2$, $MnOx/CeO_2$, Rh_2O_3/CeO_2 with Al_2O_3 or zeolit as a carrier (Fino et al., 2003, Park et al. 2009). In SCR, reaction agent is dosed before catalytic bed into flue gas stream with NH_3/NO stoicheiometric ratio up to 1.1 mol/mol (European IPPC Bureau, 2003). Nitorgen oxides are then reduced according the identical stoicheiometric equations as for SNCR.

Compared to SCR, SNCR presents lower investment and operating costs. SCR, however, is a more efficient deNOx method.

Tab. 1: Technologies for nitrogen oxides (NOx) cleaning

	Efficiency of NOx reduction [%]			source
	min	max	avg	
Selective non-catalytic reduction of NOx (SNCR)	40	70	55	European IPPC Bureau, 2003
Selective catalytic reduction of NOx (SCR) – fixed bed catalytic reactor	90	94	92	
SCR – fabric catalytic filter (e.g. Remedia®)	N/A		33.2	Dvorak et al., 2010
SCR – ceramic catalytic filter (e.g. Cerafil TopKat®)	76	80	78	Startin and Elliot, 2006

2. Increase of efficiency of deNOx secondary technologies

Due to strict emission limits and environment protection requirements, it is necessary to increase efficiency of deNOx secondary technologies. This may be achieved via installation of a new technology with higher efficiency or by supplementing the existing technology with a new one. For installation of more technologies, total deNOx efficiency is calculated using (3) relation for installation of two technologies and using (4) relation for installation of three technologies.

$$\eta_{NOx,C} = \eta_{NOx,1} + (1-\eta_{NOx,1}) \times \eta_{NOx,2} \qquad (3)$$

$$\eta_{NOx,C} = \eta_{NOx,1} + (1-\eta_{NOx,1}) \times \eta_{NOx,2} + (1-\eta_{NOx,1}) \times (1-\eta_{NOx,2}) \times \eta_{NOx,3} \qquad (4)$$

$\eta_{NOx,C}$ – total efficiency of NOx reduction [-]
$\eta_{NOx,1}$ – efficiency of NOx reduction in technology 1 [-]
$\eta_{NOx,2}$ – efficiency of NOx reduction in technology 2 [-]
$\eta_{NOx,3}$ – efficiency of NOx reduction in technology 3 [-]

Efficiency of deNOx technologies is then calculated using following relation (5):

$$\eta_{NOx,i} = \frac{x_{NOx,i-in} - x_{NOx,i-out}}{x_{NOx,i-in}} \qquad (5)$$

$\eta_{NOx,i}$ – efficiency of NOx reduction in technology i [-]
$x_{NOx,i-in}$ – inlet concentration of NOx to technology i [mg/m_N^3]
$x_{NOx,i-out}$ – outlet concentration of NOx from technology i [mg/m_N^3]

2.1 Examples of increase of efficiency of deNOx secondary technologies

Possibilities of increase of efficiency of deNOx secondary technologies may be illustrated on following example of two real municipal solid waste (MSW) incineration plants with waste processing capacity of 15 t/h. First of MSW incineration plants (A incineration plant) engages a flue gas cleaning system (Pavlas and Touš, 2009) which consists of SNCR, ESP, REMEDIA® catalytic filtration and NaOH wet flue gas cleaning (see Fig. 1).

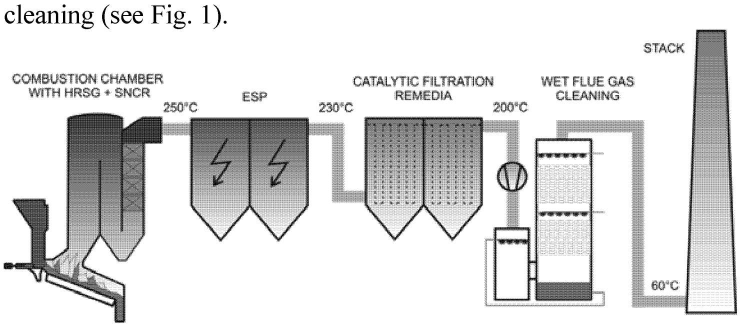

Fig. 1: Scheme of MSW incineration plant A

Second of MSW incineration plants (B incineration plant) engages a flue gas cleaning system which consists of SNCR, ESP, $Ca(OH)_2$ semi-dry flue gas scrubber, active carbon dosing and baghouse with fabric filters (see Fig. 2).

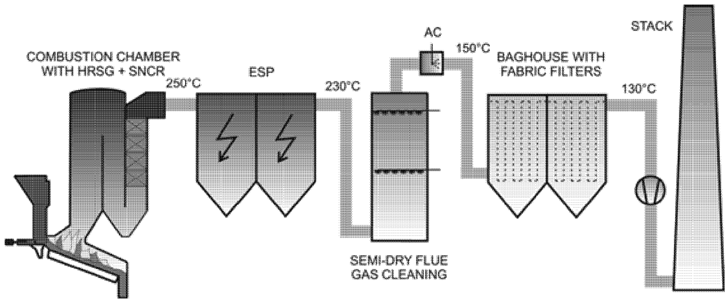

Fig. 2: Scheme of MSW incineration plant B

Both incineration plants reduce NOx using SNCR and their outlet NOx concentrations reach 170 mg NOx/m_N^3 (A incineration plant), 160 mg NOx/m_N^3 (B incineration plant) respectively. However, emissions limit 200 mg NOx/m_N^3 according to legislation (Council Directive 2000/76/EC, 2000) is exceeded often. Any more stringent legislation will result in urge to replace existing SNCR technology with SCR technology with fixed bed or by supplementing existing SNCR technology with SCR technology with catalytic filter. Maximum SNCR technology efficiency reaches 70 %, SCR technology with fixed bed efficiency reaches 94 %, combination of SNCR and SCR with catalytic filtration technologies efficiency reaches up to 80 % (SNCR + SCR Remedia®), resp. 94 % (SNCR + SCR Cerafil TopKat®). Combination of SNCR and SCR with fixed bed technologies may theoretically reach up to 98 % efficiency. NOx outlet concentrations for various technologies and their combination in relation to A incineration plant are presented in Fig. 3.

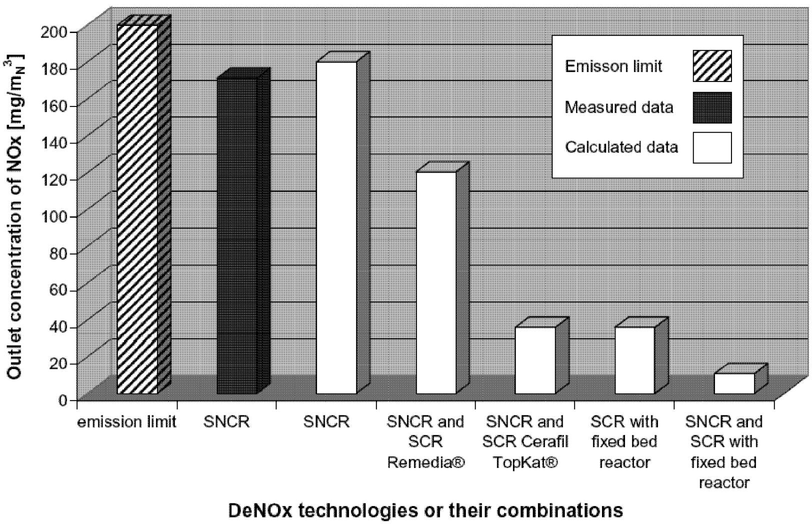

Fig. 3: Example of potential decrease of NOx concentration in flue gas by application of various technologies on MSW incineration plant A.

3. Discussion

Selection of a new technology or combination of technologies always has to be preceded by thorough financial balance and comparison of investment and operating costs. Increase of deNOx efficiency A incineration plant may be obtained by dosing of 25 % water solution of ammonia which is utilized in this plant as a reduction agent for SNCR process. Purposeful increase of solution concentration will result in NOx reduction at REMEDIA® catalytic filter which is now operated in reduction of particulate matter (PM) and polychlorinated dibenzo-p-dioxines and furans (PCDD/F). If stoicheiometric ratio of NH_3/NO before REMEDIA® catalytic filter reaches around 1.1 mol/mol, deNOx efficiency at REMEDIA® catalytic filter may reach 33.2 % and thus overall deNOx efficiency of the whole flue gas cleaning system may reach up to 80 %. Investment costs equal zero and operating costs will rise by adding extra reaction agent. Application of any other deNOx technology is irrelevant in this case.

Selection of deNOx technology is more complicated for B incineration plant. If existing baghouse with fabric filters should be replaced by SCR catalytic filters, there are two options:

- Replace existing baghouse with fabric filters with REMEDIA® filters (construction of the baghouse remains basically the same). Advantages include higher deNOx efficiency (total 80 %) and lower investment costs. Disadvantages include need to raise temperature of flue gas before catalytic filter to reaction temperature of 220 to 240°C (by direct heating of flue gas or installation of heat exchanger). Proper function is then conditioned by increase of ammonia compounds concentration before filter to ratio of NH_3/NO to 1.1 mol/mol.

- Replace the whole baghouse with fabric filters for baghouse designed for Cerafil TopKat® ceramic catalytic filters. Advantages include high deNOx efficiency (total 94 %) and possibility to place a new baghouse with ceramic filter at the spot of existing baghouse. Disadvantages include higher investment costs and need to raise temperature of flue gas to SCR reaction temperature. Proper function is then conditioned by increase of ammonia compounds concentration before filter to ratio of NH_3/NO to 1.1 mol/mol.

Obviously, it is possible to install SCR technology with fixed bed. Advantages include installation of SCR technology beyond ESP and thus flue gas temperature increase is eliminated. High deNOx efficiency (94 %) is also a positive effect. However, disadvantages include high investment costs; need to find a proper location for new SCR technology with fixed bed in current technological arrangement and sensitivity to high concentrations of acidic components (SO_2, HCl a HF) – in the case with SCR technology with fixed bed before semi-dry flue gas cleaning.

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