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NOx Reduction - The Challenge for Innovative Concepts in Europe

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ABSTRACT

During combustion, most of the waste's nitrogen content is transferred to the flue gases as nitrogen oxide, NO_x. The EU Waste Incineration Directive defines a maximum emission limit value for NO_x of 200 mg/Nm^3 as a daily average value referred to 11% O_2. Based on National Emission Ceilings (NEC) defined by the Gothenburg Protocol, it can be expected that the limit values for NO_x in the EU will become even more stringent. In some European countries (e.g. The Netherlands, Austria, Switzerland) a lower emission limit has already been introduced.

Selective Catalytic Reduction (SCR) technologies are used in many cases to achieve the above-mentioned NO_x limits. However, there are drawbacks to SCR systems such as high investment cost. Operation cost is also quite high due to the energy consumption necessary for the reheating of the flue gas as well as the increased pressure loss. Innovative technologies are therefore required to make it possible to reconcile both requirements: reduced emissions and increased energy efficiency.

Selective Non-Catalytic Reduction (SNCR) systems are based on the selective reaction of ammonia or urea injected into the upper furnace. In many cases SNCR technologies are limited by the ammonia slip which increases in case of more stringent NO_x requirements. According to the relevant BREF document, an ammonia slip limit of 10 mg/Nm^3 is generally required at the stack. In order to achieve reduced NO_x values, it is necessary to implement measures to reduce ammonia slip, by means of either a wet scrubber or a High-dust catalytic converter. EfW plants in Mainz (Germany) and Brescia (Italy) are examples of operational plants combining SNCR with such a catalytic converter type.

In addition R&D activities are carried out on the development of simplified reaction mechanisms to be implemented in Computational Fluid Dynamics (CFD) codes. With these tools it will be possible to describe the interaction between turbulent mixing, radiation and chemical reaction rates.

Another option to achieve low NO_x values (below 100 mg/Nm^3) is the reduction of NO_x by so-called primary measures, e.g. the Very Low NO_x process (VLN), which has been developed by MARTIN jointly with its cooperation partners. The VLN process is based on a grate-based combustion system. The "VLN gas" is drawn off at the rear end of the grate and is reintroduced into the upper furnace in the vicinity of the SNCR injection positions. NO_x will be reduced significantly, ensuring low NO_x emission values at the stack as required, at low values for ammonia slip. The new EfW plant in Honolulu (USA) will be equipped with the VLN process. In Coburg (Germany), the VLN process will be retrofitted in an existing installation.

This paper documents the potential and the limitations of different measures for NO_x reduction as well as examples of recent innovative EfW plants in Europe using MARTIN technologies successfully.

Keywords: NO_x reduction, Selective Catalytic Reduction (SCR), Selective Non-Catalytic Reduction (SNCR), catalytic converter

1. INTRODUCTION

Sustainable urban development needs a reliable supply of heat and power as well as efficient treatment of Municipal Solid Waste (MSW). The emissions from Energy from Waste plants (EfW plants) are strictly limited by the EU Incineration Directive, which sets the limit on NO_x emissions to 200 mg/Nm^3. Moreover the need for further improvement is reflected by new German legislation in effect, requiring 100 mg/Nm^3. In the Netherlands, there has long been a limit of 80 mg/Nm^3. Other European countries are expected to follow due to the national emission ceilings stipulated in the Gothenburg protocol and the resultant EU directive 2001/81/EC [11]. Some European countries restrict the emission of ammonia slip, for instance in Austria and Switzerland where the emission of ammonia is restricted to 5 mg/Nm^3. Even if the ammonia slip is not restricted by regulations it should not be higher than 5-10 mg/Nm^3 as ammonia may cause an odour of the flue gas residues [13].

Selective Catalytic Reduction (SCR) technologies were used in most cases to achieve the above-mentioned NO_x limits. However, there are drawbacks to SCR systems such as high investment costs and the energy consumption necessitated by the reheating of flue gas as well as the increased pressure loss. Consequently, SCR falls short of the efforts to increase energy efficiency encouraged by the EU regulations. New technologies are therefore required to make it possible to reconcile both requirements: reduced emissions and increased energy efficiency.

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2. FORMATION OF NO$_x$ BY GRATE-BASED COMBUSTION TECHNOLOGY

NO$_x$ is the generic term for the group of nitrogen oxides. Essentially the oxides emitted during the combustion of MSW are nitric oxide (NO), nitrogen dioxide (NO$_2$) and nitrous oxide N$_2$O. NO$_2$ is mainly formed after emission into the atmosphere by the oxidation of NO with the air oxygen or ozone (O$_3$). This formation occurs slowly and plays a minor role in the hot flue gas. However more than 95% of nitrogen oxides are emitted in form of nitric oxide NO [1] and is converted in the atmosphere to NO$_2$.

2.1 Formation mechanisms

The NO formation in the combustion chamber is a very complex process. According to the nitrogen source and the reaction medium it is differentiated into three mechanisms (figure 1):

- Thermal NO formation
- Prompt NO formation
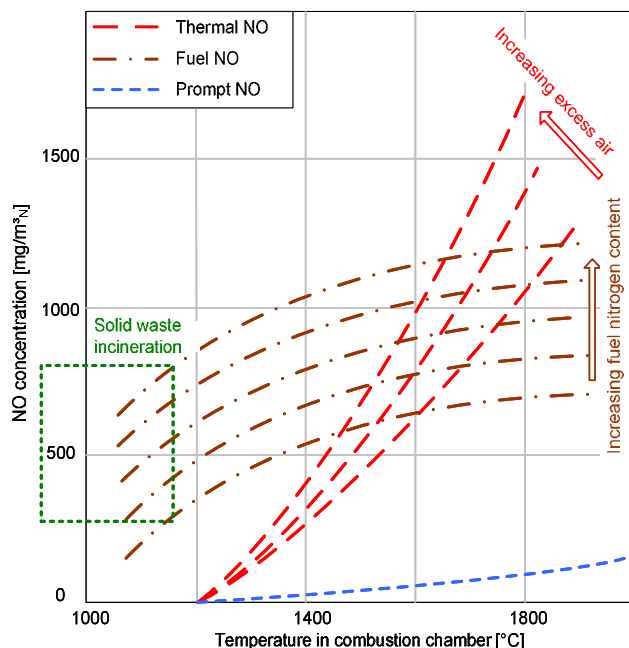- Fuel NO (NO formation from fuel nitrogen)

Fig.1   NO$_x$ formation in dependence on temperature
[Prof. Quicker]

"Thermal NO" is produced by oxidation of atmospheric molecular nitrogen and occurs significantly at very high temperatures above 1,500°C [2].

"Prompt NO" appears in the area of the flame front (reaction zone) under fuel-rich conditions. It is however generally accepted that prompt NO represents an insignificant part in the overall NO formation, especially for the combustion of solid fuels like coal or waste [3].

If the fuel contains chemically bound nitrogen in the form of organic (e.g. amine, amide, nitrile) or inorganic nitrogen compounds (e.g., ammonia or HCN), NO will be emitted during the combustion process according to the "Fuel NO" mechanism. Under the usual conditions of temperatures, air distribution and MSW composition, the Fuel NO mecha-

nism plays the most important role in the formation of NO$_x$ in the case of grate-based waste combustion technology [4].

2.2 Fuel NO mechanism

MSW (dry matter) contains on average 1% nitrogen [5]. Proteins contribute to over 30% of the whole N amount in waste, polyamides represent approximately 20%, polyurethane less than 10%, and a bigger amount of resins such as urea and melanin resin contribute too.

The formation of NO from fuel nitrogen occurs in two different reaction paths, as homogeneous gas phase reactions or as heterogeneous reactions involving the char surface.

First of all the nitrogen bound in the fuel is partially released during the volatilization phase. The distribution of nitrogen between volatiles and char, the volatilization rate, as well as the composition of the volatile nitrogen depend on fuel structure, temperature, heating velocity and residence time. For coal it could be shown that nitrogen is released mainly as HCN. However, in the case of biomass mainly NH$_3$ is released. The release takes place at much lower temperatures than with coal. Similar results also apply to MSW [6]. The nitrogen compounds released as primary products afterwards undergo a multiplicity of chemical reactions in the gas phase to form NO and N$_2$. Because nearly the entire nitrogen is released primarily as NH$_3$, HCN and NO, only these primary products are considered for the reaction kinetics in the gas phase [6]. Here are only indicated the most important reactions from a multiplicity of formation and degradation reaction pathways. Indeed the detailed NO mechanism of Glarborg for the hydrocarbon/nitric oxide interactions contains, e.g. 438 reactions and more than 100 species [7].

Figure 2 summarizes the most important reactions steps of the Fuel NO formation process.
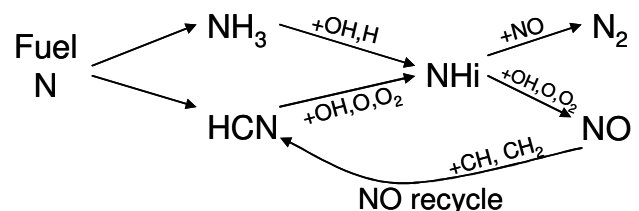

Fig.2   Most important partial steps of the Fuel NO formation [6]

NH$_3$ and HCN are released first from the fuel. From it either N$_2$ or NO is formed with NHi as an intermediate product according to the combustion conditions. The NO can again be degraded through the NO recycle process (in the case of fuel-staged combustion).

In summary the following parameters have a particular influence on the formation of Fuel NO:

- Nitrogen content of the fuel
- Characteristics of the nitrogen bond with the fuel
- Combustion conditions (e.g. temperature, air distribution) particularly oxygen availability in the pyrolysis and gasification zone, as well as residence time under reducing conditions

3. NO$_x$ Reduction Processes

NO$_x$ abatement is generally divided into two categories: "Primary Measures" involve treatment prior to or during the combustion process. Generally there is a wide range of primary reduction techniques, such as staged air combustion, reduced excess air, reburning, flue gas recirculation and combustion with pure oxygen. Primary measures can achieve up to 70% reduction [8] but "Secondary Measures" or post-combustion measures are often necessary to comply with the NO$_x$ emission regulations. Examples of secondary measures are the Selective Non-Catalytic Reduction (SNCR) or the Selective Catalytic Reduction (SCR). The positions of these processes are shown in figure 3.
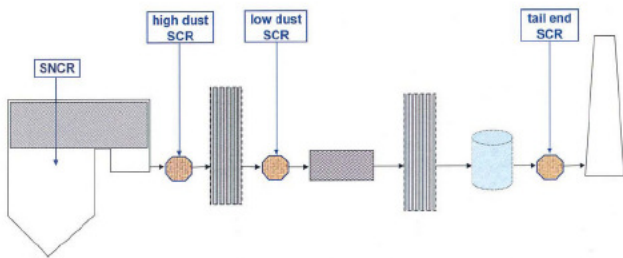

Fig. 3 Positions of secondary measures for NO$_x$ reduction [16]

3.1 Very Low NO$_x$ (VLN)

In 1998 already MARTIN together with institutes developed and optimized successfully a multi-staged process in a pilot plant which consists of gasification with air on the MARTIN reverse-acting grate ($\lambda = 0.4 - 0.8$) followed by an independent stoichiometric post-combustion of the gases generated ($\lambda = 1.2 - 1.4$) as a primary measure for the NO$_x$ reduction [21, 22]. Various studies and tests in particular position / distribution of overfire air nozzles and mixing possibilities of the uncombusted gases in the post-com

bustion zone followed in pilot- and large-scale.

In 2008 MARTIN and its cooperation partners developed the Very Low NO$_x$ (VLN) process as a large-scale primary measure, which achieves much lower NO$_x$ emissions than the conventional SNCR process without energy losses [12].

The VLN process (figure 4) is based on a classical grate-based combustion system for municipal waste, where the so-called "VLN gas" is drawn off at the rear end of the grate and is reintroduced into the upper furnace just below the ammonia injection positions. The positive effect of this patented process is twofold: On the one hand, drawing off the VLN gas leads to combustion conditions which promote the inherent NO$_x$ reduction processes such that Fuel NO$_x$ is to a large extent reduced to nitrogen. On the other hand, the reinjection of the VLN gas cools the flue gases down and enforces their mixing with injected ammonia or urea. This leads to improved efficiency of the SNCR system.

NO$_x$ values of 80 mg/Nm^3 with an NH$_3$ slip of less than 10 mg/Nm^3 have been reached in tests at the EfW plant in Bristol/USA. Further industrial-scale tests in Thiverval/France and Oita/Japan have confirmed these results. At the Thiverval plant, which has a MSW throughput of 12 t/h, NO$_x$ could be reduced from 190 mg/Nm^3 to 80 mg/Nm^3 during test operation with the VLN components.

The VLN gas has a temperature below 300°C and is re-injected at a position at which the furnace temperature is around 1,000°C. This typically corresponds to a level of 8 to 12 m above the grate depending on the capacity of the unit and the type of waste. The overfire air pressures are reduced to around 10 mbar, which is considerably less than in conventional EfW plant design. Nevertheless, super-stoichiometric conditions are reached at the overfire air level, which is an advantage compared to air-staged or fuel-staged combustion systems. The residence time from the last combustion air injection at the overfire air level to the 850°C level in the furnace is significantly increased. A further advantage of the VLN system is the reduced flue gas velocity in the lower furnace due to internal recirculation via the VLN duct. This leads to a reduction in the fly ash carried over to the boiler.
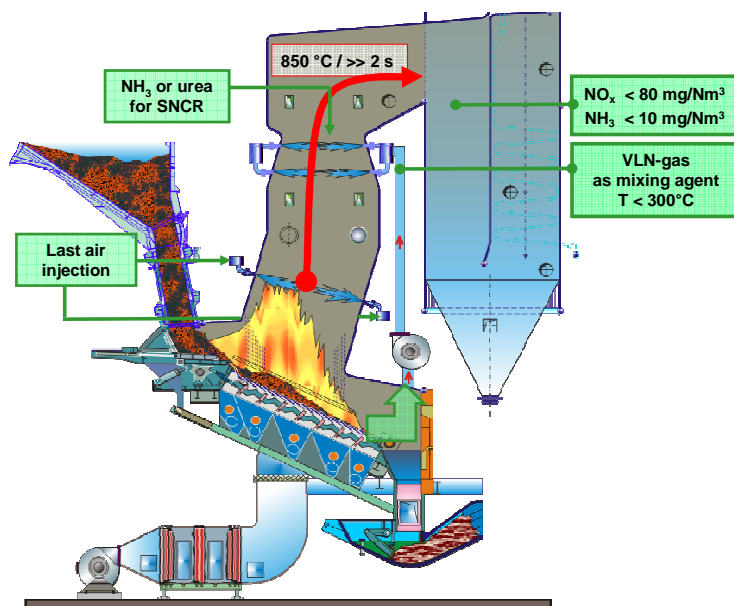

Fig. 4 VLN process (NO$_x$ / NH$_3$ referred to 11% O$_2$)

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The VLN gas is re-injected into the front and rear side of the narrow section of the upper furnace. This leads to an intensive barrier of turbulence, which reduces the flue gas temperature and blocks the passage of flames or unreacted gases. The test plant results give rise to the expectation that corrosion is significantly reduced in the furnace above the VLN level as well as in the superheaters. On the other hand, temperatures between the overfire air and the VLN level are higher than in conventional combustion and higher grades of furnace protection material should be used there. Another advantage of the VLN system is the reduced excess air rate, which allows cost reduction in the boiler and flue gas cleaning and improved boiler efficiency. The new EfW plant in Honolulu (USA) will be equipped with the VLN process. In Coburg (Germany), the VLN process will be retrofitted in the existing installation.

3.2 Selective Non-Catalytic Reduction (SNCR)

SNCR is a widespread standard process for the reduction of NO_x emissions. This process involves the selective gas phase reaction of nitrogen-containing reducing agents (ammonia NH_3, urea $(NH_2)_2CO$) with NO_x in the presence of oxygen to form primarily nitrogen and water. These reactions occur in the temperature range between approx. 850°C to 1,050°C. The major process parameters impacting SNCR performance include:

- Type of reducing agent
- Sufficient stoichiometry of reducing agent
- Temperature window
- Efficient mixing of reducing agent with flue gases
- Sufficient retention time

In an ideal case of mixing and reactivity, the best results could be attained with a stoichiometry of 1. In practice only 50 - 60 % of NO_x reduction is reached by the addition of ammonia or other reducing agents at a stoichiometry of 2 [9]. Laboratory experiments show that retention times of 50 ms are sufficient for the SNCR reaction. In practice it is necessary to obtain retention times at suitable temperatures of up to 1 s in order to reach reasonable reduction rates. This is mainly due to the fact that the mixing and turbulence is insufficient in combustion furnaces.

MARTIN has developed a process based on SNCR (figure 5). The nitrogen oxides produced during combustion are reduced by injecting the reducing agent into the furnace. A second mass flow is required in order to achieve a uniformly fine distribution. Compressed air, steam or softened fresh water are used for this purpose. The two mass flows are combined in a mixing area directly at the nozzle head. This results in low deadtimes for control of the ammonia mass flow as well as optimized consumption and low ammonia slip (NH_3) in the flue gas. Further optimization is possible using an in-situ NH_3 measuring device. The control system ensures that injection always takes place in the optimum temperature range as a function of the current furnace temperature. MARTIN has installed this SNCR system successfully in various plants in Europe. Guaranteed NO_x values of up to 70 mg/Nm^3 are reliably achieved with a low level of NH_3 slip at the boiler outlet. Significant advantages of an SNCR process are:

- Simple handling
- Stable operation
- Attainable lower NH_3 slip

The $NO_{x\ stack}$ -$NO_{x\ boiler\ outlet}$-$NH_{3\ boiler\ outlet}$ control concept (which includes a balance control for smooth shifting of the NH_4OH injection between both injection levels) is implemented for demand-oriented injection and optimal use of aqueous ammonia given varying NO_x raw gas concentrations and combustion conditions.
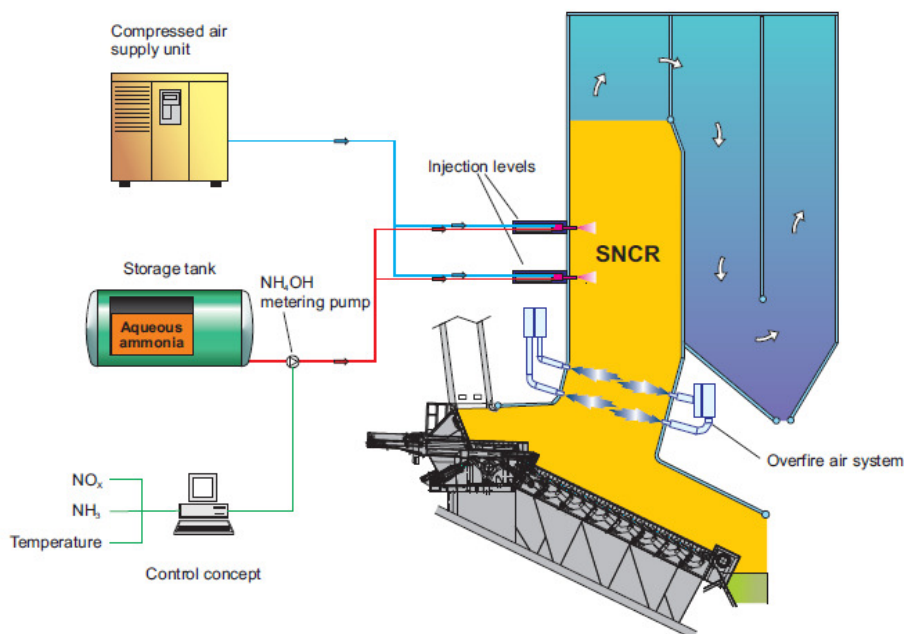

Fig. 5 MARTIN Compressed Air SNCR System

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3.3 SNCR combined with a High-dust Catalytic Converter

In many cases SNCR technologies are limited by the ammonia slip which increases in case of severe NO_x requirements. According to the BREF, a limit of 10 mg/Nm^3 is generally required at the stack [10]. In order to achieve reduced NO_x values of 70 - 100 mg/Nm^3, it is necessary to implement measures to reduce the ammonia slip by means of either an acidic wet scrubber (washing out of ammonia) or a high-dust catalytic converter. The catalyst type and the pitch must be optimized for using a catalytic converter under such conditions (figure 6). More details about catalytic converters are described in 3.4.
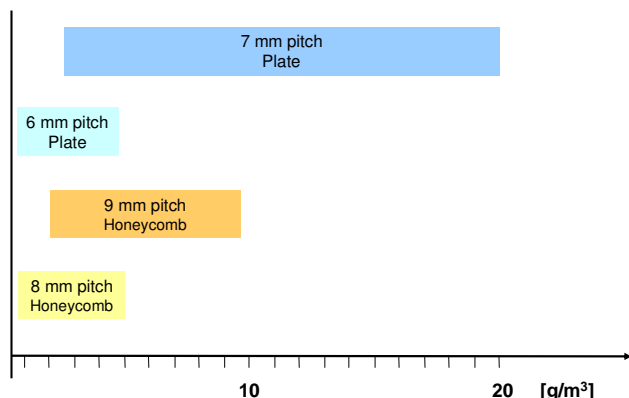

Fig. 6 Pitch and catalyst type in dependence on dust content [based on 17]

EfW plants in Mainz (Germany) and Brescia (Italy, figure 7) are examples of operational plants combining SNCR with such a high-dust catalytic converter type.
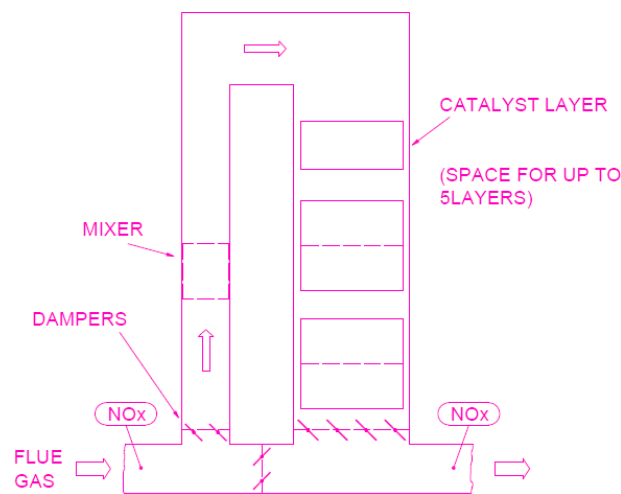

Fig. 7 High-dust SCR layout (EfW plant Brescia) [19]

In the EfW plant in Brescia the following results are be achieved with aqueous ammonia 25 % and a negligible pressure loss [19]:

NO_x	80-90 mg/Nm^3	→ 60-70 mg/Nm^3
NH_3 slip	10-30 mg/Nm^3	→ 2-6 mg/Nm^3
NH_3 consumption	0.22 m^3/h	→ 0.18 m^3/h

Much higher energy efficiency (no need of gas reheating, small gas pressure losses, low impact on fan consumption),

a simpler installation, an improvement of the reaction between ammonia and NO_x and finally lower investment and operating costs are the essential advantages. However, erosion, clogging and poisoning of the catalysts decrease their life-time. For the reduction of the erosion modifications were implemented:

- New geometry in order to increase plates stiffness
- Increased plates thickness
- New composition of catalytic material to increase plates hardness

However, although the life-time of the catalysts is shorter the costs (catalysts versus energy (SCR) and ammonia consumption (SNCR)) are relatively low in comparison to other SCR processes [19].

3.4 Selective Catalytic Reduction (SCR)

The same reduction mechanisms are used in both the SCR (Selective Catalytic Reduction) process and SNCR technology. The catalyst for SCR is made of platinum, rhodium, titanium oxide, vanadium pentoxide and other materials [20]. Due to the catalyst the reaction can take place at a lower temperature normally around 250°C, however, references in the temperature interval between 180°C to 350°C are available [13].

More catalyst material is required for temperatures below 250°C and there is an increased risk of damages due to adhesions and catalyst poisoning. There are three different possibilities for arranging the catalytic converter (figure 3):

- High Dust
- Low Dust
- Tail End

In the "High Dust" process the catalyst block is located directly at the boiler outlet: there is no downstream dedusting or flue-gas desulfurization (FGD). The downside of this switching is that the service life of the catalytic converter is shortened due to clogging and deactivation.

In the "Low Dust" process the catalytic converter is situated downstream of the electrostatic precipitator but upstream of the FGD. The advantage here is the reduced dust burden; however, flue gas desulfurization has not yet taken place which is potentially a disadvantage.

In the "Tail End" process the SCR catalytic converter is located at the end of the flue gas cleaning process thus minimizing negative influences. The cost of reheating the flue gases downstream of the FGD to the required SCR temperature (by approx. 60°C) is one drawback of this variant. For this purpose a heat exchanger is used to transfer the heat from the flue gas coming from the DENOX unit to that coming from the FGD unit. Additional burners are often needed in order to ensure that the required temperatures are reached, which in turn increases cost.

The dust and heavy metal content of the flue gas responsible for clogging and deactivating the catalyst are the determining factors with regard to selecting one of the three proposed processes. Consequently, the flue gas cleaning system is specially tailored to each EfW plant.

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A significant advantage of the SCR process is the high NO reduction rate of 90% at virtually stoichiometric conditions. As a result, the specific ammonia requirement is lower than that of comparable SNCR processes. Nevertheless, this advantage is overshadowed by numerous disadvantages:

- Energy consumption for the reheat (and electricity consumption for the increased pressure drop)
- High cost of investment and operation
- Limited lifetime of the catalytic material
- Additional pressure drop in the flue gas stream

A comparison between the SNCR process and two variants of SCR technology with respect to costs reveals that the investment costs for a SCNR system are less than a third of comparable costs for a SCR unit. In terms of total annual cost, SNCR technology is 50 % cheaper than a SCR unit with "Tail-End" despite higher operating costs [20].

Contrary to the SNCR process ammonia has to be added to the process in a small surplus only. For reducing the amount of NO_x with 80 % some few percent (3-5 %) ammonia more than the stoichiometric amount has to be added compared to 2,5 times for the SNCR [13].

4. Simulation of NO_x Reduction by CFD

Today's research focuses on the development of simplified reaction mechanisms to be implemented in Computational Fluid Dynamics (CFD) codes. With these tools it will be possible to describe the interaction between turbulent mixing, radiation and chemical reaction rates.

Calculations based on CFD simulation are extremely suitable for implementing simple, fast and cost-effective parameter studies. In this way, the formation and reduction of nitrogen oxides in EfW plants can be calculated numerically. In addition, important influencing parameters can be determined and subsequently implemented in real plants. By MARTIN in cooperation with the Technical University of Munich (Germany) studies were done including boundary conditions and measurement values from a large-scale EfW plant and integrating these in a CFD calculation model. The nitrogen oxides in EfW plants originate almost entirely from the nitrogen contained in the fuel, as a result of which particular attention has to be paid to simulation of the Fuel NO. Based on a convergent solution for the simulation of NO formation using an NO_2 starting concentration of 400 mg/Nm^3, calculations were made for nitrogen oxide reduction using the SNCR process. Several calculation cases with different injection media and nozzle arrangements were examined. It was confirmed that the temperature distribution and carrier medium are key in terms of SNCR process effectiveness. A higher NO_x removal rate (64 %) with lower NH_3 slip (1.1 mg/Nm^3) was achieved by using compressed air instead of water to atomize the aqueous ammonia. The results obtained are a good reflection of the measurements made during and experiences gained from real plant operation [20].

Together with universities and institutes MARTIN continues these studies of NO_x formation and reduction processes by using CFD including waste-bed modelling for creating starting values.

5. CONCLUSIONS

EfW plants have attained a high technological standard. In the EU, emissions from EfW plants are largely reduced by applying the Waste Incineration Directive with its limit values for dioxins (PCDD/F), dust, NO_x and many other pollutants.

The need for further improvement is reflected by new legislation. On the one hand, more stringent NO_x values may become compulsory due to the NECs of the Gothenburg protocol. On the other hand, an increase in energy efficiency will be encouraged by the Waste Framework Directive [15]. This is why there is a need for new technologies that make it possible to reconcile both requirements: reduced emissions and increased energy efficiency. One promising example is the VLN technology developed by MARTIN jointly with its cooperation partners.

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