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Reduction of NO_X at New Mobile Experiment Unit Using Catalytic Filtration

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With the growing proportion of thermally removal waste relate tightening of emission limits for exhaustion of pollutants into the atmosphere. It is therefore necessary to come up with new, more effective but also economically acceptable methods. Current secondary methods for disposal of hazardous pollutants contained in flue gas mostly require the installation of separate equipment for each of the controlled pollutants. To remove solid pollutants (ashes) are used fabric filters, ESP (electrostatic precipitator) or cyclones. The destruction of NO_X is used SNCR (selective non-catalytic reduction) or SCR (Selective Catalytic Reduction). Acid components of flue gas (SO2, HCl, HF) is often removed dry, semi-dry or wet scrubber, VOC and PCDD/ F are removal such as using activated carbon adsorbers, etc.

The article deals with the selective catalytic reduction of nitrogen oxides at fabric and ceramic filters with v implemented catalyst. Passing flue gas through the wall of filter is also to remove solid pollutants. This combination can be termed as a catalytic filtration. Used catalysts also allow decomposition of PCDD/ F at an acceptable pressure drop. Objective research is primarily overall evaluation filtration materials, evaluation of economic aspects and comparison with current methods of NO_X reduction.

To verify and quantify these assumptions have been built the experimental unit capable to simulate of operating conditions occurring in real plants.

1. Introduction

Commonly used methods for secondary NO_X reduction (SCR, SNCR) are based on the reaction of NO and NO_2 contained in the flue gas with a reducing agent based on ammonia (urea, ammonia water, ammonia, and others) in the presence of a catalyst. Used catalysts are precious metals for temperatures 177-288 °C, V_2O_5/TiO_2 for temperatures 260-427 °C or zeolites in the temperature range 455-594 °C (Schnelle and Brown, 2001). NH_3/NO molar ratio ranges from 0.8 to 1.1 (European IPPC Bureau). Ongoing reactions during SCR NO_X when using NH3 as a reducing agent (Schnelle and Brown, 2001):

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

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

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Catalytic filtration is a form of SCR, where contact of pollutants and catalyst appears when a mixture of polluted gas with a reducing agent is passing through a porous wall of filter.

Currently, experiments are under way with two types of filters (fabric and ceramic). In the case of fabric filter is the catalyst V_2O_5-WO_3/TiO_2 placed between two membranes (solution Remedia ® fabric filtration). Benefits of this solution are low pressure drop, high efficiency of separation of solid pollutants and PCDD/F, it don´t absorb water, resists UV radiation, mechanical damage and temperatures up to 250 °C. The second tested material is ceramic filter - candle (solution Cerafil TopKat ® ceramic filtration). Candle reproduces the benefits of fabric filter, and above that, it can operate at higher temperatures (SCR up to 450 °C). Ceramics is also resistant to significant fluctuations in temperature and satisfies the conditions for wider use (Startin and Elliot, 2010). The catalyst is implemented into the structure of the candle.

2. Experimental Unit

There were projected and constructed a new unique mobile experimental unit called INTEQ II (see Fig.1 and Fig.2). Project was based on experiences from previous research and operating at unit called INTEQ I (Dvořák, et al., 2010).

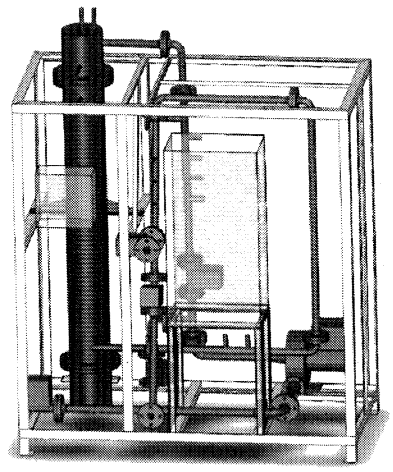

Figure 1: INTEQ II – model

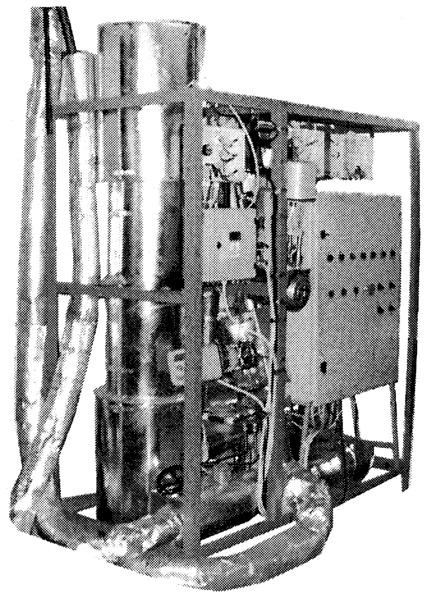

Figure 2: INTEQ II – real situation

There were carried basic experiments of above mentioned fabric filter. The reduction efficiency reached 33.2% in the stoichiometric ratio NH_3/NO = 1.1 mol/mol. In tests with non-standard over-stoichiometric ratio the efficiency decreased to 4.5% due to unreacted ammonia compounds, mainly $(NH_4)_2SO_4$. Filter was not damaged and after regeneration by water extraction, there was measured the efficiency of reduction in the original values. When the unit was operated in real plant, it was destroyed by aggressive substances in the flue gas. Therefore, during design of new unit was necessary to avoid these problems by satisfying the following requirements (Dvorak et al. 2010):

- Constant temperature, min. 220°C
- Construction is made from resistant material
- Automatic dosing of ammonia in selected molar ratio, thereby preventing ammonia slip and the formation of hazardous compounds

The new unit is compact, easily removable and transportable. It is also possible to test not only different filters in form of bags, but also pieces of fabric. The main operating parameters of both units are showed in Tab. 1.

Table 1: Comparison of operating parameters

Parameters	INTEQ I	INTEQ II fabric filter	INTEQ II ceramic filter
Real flow [m^3/h]	1000	28.65	13.39
Number of bags	15	1	1
Operation temperature [°C]	250	250	360
Filtration rate [m/min]	0.93	0.8-1	0.8-1

The principle of operation of the unit can be seen on the technological diagram (see Fig.3). The inlet gas stream can be dosed reductant and if necessary, also NO to increase its concentration. If flue gas does not have the desired temperature it can be heated by using installed electric heating. To regulate the temperature and gas flow are installed automatically controlled valves. Pipeline includes sensors to measure pressure, temperature and gas flow, which in cooperation with the operation system provides suitable reaction conditions. Before and after the filtration chamber are installed gas analyzers with logging. It enables automatic dosing reductant (or also a NO), depending on the concentration of pollutants in the flue gas. The fan produces a maximum vacuum 4.9 kPa. To maintain an acceptable pressure drop is installed pulse regeneration filters.

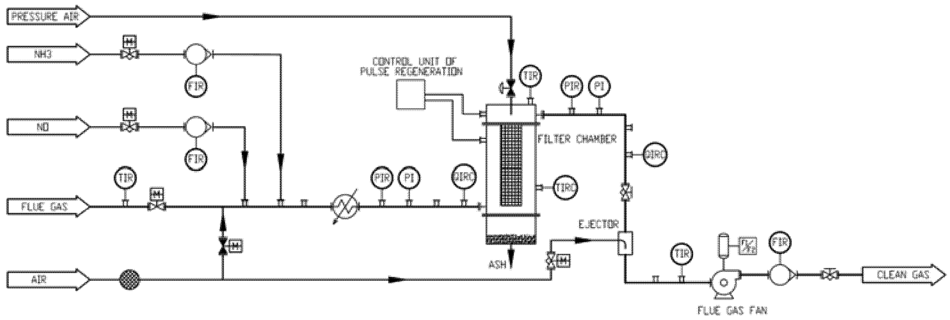

Figure 3: Technological scheme

3. Experiment in Operating Conditions

To get another perspective on the issue of NO_X reduction and to obtain operational data, an experiment was carried out in the waste incinerator in Liberec, Termizo. In the incinerator was installed Remedia fabric filter for separating solid pollutants and dioxin degradation. NO_X reduction is normally provided by SNCR with NH_4OH as a reduction agent. The manufacturing output of the incinerator is 12 t/h and gas flow in the chimney is approximately 79,000 m_N^3/h. The temperature in the combustion chamber is 920-925°C and temperature at the dioxin filter is about 222 °C (Bébar, et al., 2010*)*. When replacing the sleeves due to the end of their durability, we carried out experiment with an increased dosage of the reducing agent for SNCR. Ammonia slip was assumed and so, the reduction agent could get up to the dioxin filter. There could therefore lead to possible SCR NO_X. The results obtained should be taken as very approximate, but they can be considered valuable for further research.

For summary of measured results see Fig.4. Under normal operating conditions, molar ratio NH_3/NO before the SNCR ranges from 1.4 to 1.6 and NO_X reduction reaches 50 - 60% (measured at the exhaust end). Increasing the molar ratio to about 3.25 NH_3/NO, NO_X reduction increased up to 80-85%. These results can not accurately determine effect of the reduction of NO_X at fabric filter Remedia due to reagent dosing before SNCR and relatively inaccurate measurement of the dosed quantity. Therefore, it can be only assumed, that is reduction actually achieved by catalytic filters. On the other hand, an increase of reduction of about 30% would respond with the results shown in (Dvorak et al. 2010). It is necessary to bear in mind, that the NO_X reduction and decomposition of PCDD/F are competitive reactions. Therefore we need to find the optimum ratio, which is one of the research goals.

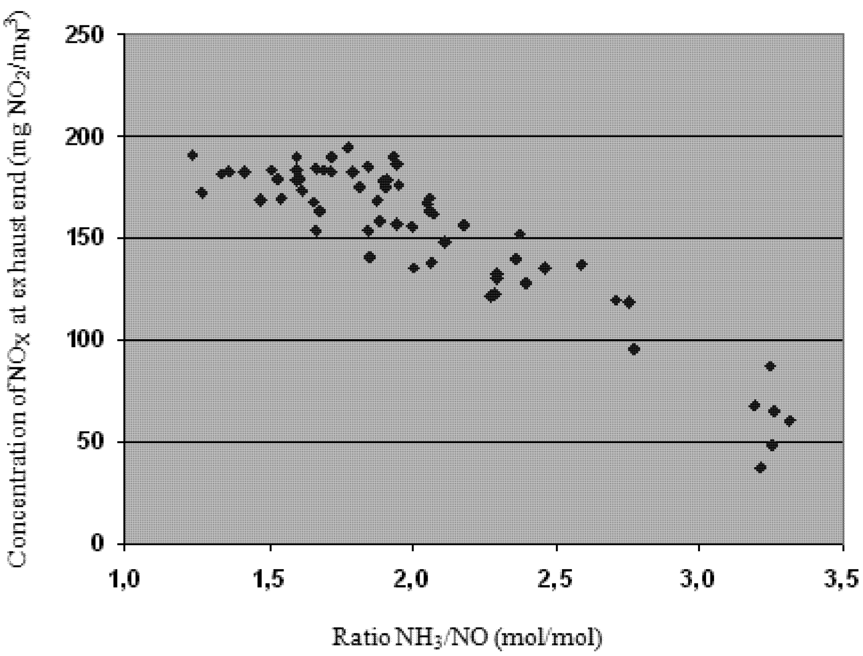

Figure 4: Influence of ratio NH_3/NO before SNCR on realized residual content of NO_2 at exhaust end. (Bébar, et al., 2010)

4. Discussion

For a perfect description of the processes taking place in the catalytic filter of the above-mentioned materials is planned series of measurements. The first tests will take place in the laboratory with artificial dosing NO. Subsequently, the unit will be moved into real operation. The goal of the research is complete description of material behavior, efficiency in a wider temperature window, effect of gas composition, influence of the concentration of pollutants and reducing agent, dependence of pressure loss in the filtration rate, etc. Finally, we will try to verify and refine the predictive model describing SCR NO_X in a catalytic filter.

Results will be compiled and evaluated, focusing primarily on the economic aspect. Proportion of investments and operating costs will be quantified and will be compared with other commonly used NOX reduction technologies. Similar results as in (Dvorak et al., 2010) can be expected for fabric filter. If efficiency is about 35%, this technology may be used in combination with SNCR. Then we can safely meet tighter emission limits. Higher efficiency up to 80%, but higher pressure loss can be expected at ceramic filter (Startin and Elliot, 2010).

Acknowledgments

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