LIMITATIONS OF NO_X REMOVAL BY PULSED CORONA REACTORS

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1. INTRODUCTION

One of the obstacles to use a pulsed corona discharge in industrial applications is that NO_x concentration below a level of 1 ppm cannot be removed. Streamer discharges (pulsed corona discharges and dielectric barrier discharges) produce NO_x themselves, and the amount of NO_x depends on the deposited energy. There are presently only a few papers investigating this problem [1,2]. The authors of [1] suggested covering the electrodes with the photocatalyst TiO_2 to remove NO_x concentrations below 5 ppm. The NO_x removal efficiency with or without photocatalyst varied by not more than 10 % on a total removal efficiency of 30%.

In [2], the NO_x input of 30 ppm into the reactor was tested on semi-industrial scale, and the tests were accompanied by computer simulations, to illustrate the analysis of the reactor process and to test the applicability to traffic tunnel cleaning. It was shown that the ([NO]+[NO_2]) concentration can be reduced to a few ppm. Nitric acids are formed as main oxidation products. To reduce the acids concentration it was suggested to spray water into the discharge chamber.

In the present paper, NO_x production and removal at a low level of NO concentration in air in a pulsed corona reactor are studied. A model of the cleaning process is successfully compared with experiments; it identifies the main plasma-chemical reactions and predicts that NO_x removal can be improved by adding hydrocarbons.

2. RESULTS AND DISCUSSION

2.1. Experiments

The setup [3] has 16 parallel wire-cylinder reactors with a total volume of 322 L. It is powered by pulses of 80 kV with 15 ns rise time, 150 ns width (power) and energy per pulse of 4.3 J. The pulse repetition rate is varied from 0 to 500 Hz to set the energy density between 0 and ca. 20 J/L. The reactor is equipped with a scrubbing system. An array of venturi nozzles on top of the reactors sprays water in the corona cylinders. The water is collected and recycled. The water flow is circa 20 L/hour. The pH was varied between 8 and 11. Pulsed power is measured using the differentiating/integrating system, which is based on differentiating sensors and integrating detection [4]. Together with other design rules this ensures proper EMC (electro-magnetic compatibility) [4]. The NO_x levels are measured with Airpointer (Recordum Austria) chemoluminescence detector. We expect that the Airpointer also responds to HNO_3, HNO_2, N_2O and N_2O_5. Ozone, produced by pulsed corona, was removed to below 500 ppb in a heated borosilicate glass tube (350 C) before entering the NO_x detector. The tests were performed using a forced flow through the reactor and an addition of ca. 1 ppm NO by a controlled flow from 50 L/200 bar cylinder of N_2 with 1000 ppm of NO.

2.2. Modeling and results

To describe the removal process we used our chemical kinetics model which takes into account the non-uniform distribution of the initially activated components just after the streamer ionization front has passed [5]. These initial densities of excited molecules, atoms, radicals, ions and electrons are calculated with a Monte-Carlo particle model for planar streamer fronts [6], where the maximum electric field at the streamer head is taken as 100 kV/cm.

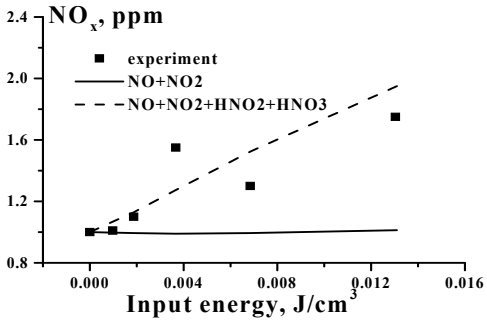

Fig. 1: NO_x removal by corona discharges in humid air.

In the figure, experimental and calculation results are compared for air with 100% humidity and $[NO]_0$=1 ppm. The agreement with experiment is better when significant components such as nitrogen oxides and nitrogen-containing acids are included. The value of $[N_2O_5]$ is almost zero. $[N_2O]$ is lower than 0.2 ppm for E=0.0131 J/cm^3. In the corona discharge, OH, H, N, and O radicals are produced in each pulse. NO is produced mainly in the reactions $O_2 + N \Rightarrow O + NO$ and $OH + N \Rightarrow H + NO$. In humid air when $[NO] \sim 0$, NO_2 is produced by the reactions $OH + HNO_2 \Rightarrow H_2O + NO_2$, and $O_2 + HNO \Rightarrow OH + NO_2$, and $[NO_2]$ decreases mainly in the reaction $OH + NO_2 + M \Rightarrow HNO_3 + M$. One source for acids is hydrated ions. The simulation shows that adding a small amount of C_2H_4 and C_3H_6 enhances the NO_x removal efficiency and decreases the concentration of acids.

CONCLUSIONS

Sub-ppm NO_x removal by pulsed corona encounters a number of serious difficulties. In the low ppm range a pulsed corona NO_x removal is balanced by a pulsed corona NO_x production. Hence, the removal process quenches below the 1 ppm NO_x level. Secondly, the chemoluminiscence measuring principle for NO_x detection also responds to nitrous oxides other than NO and NO_2, and acids. Therefore, although NO_2 has been converted to HNO_3, the achieved NO_2 removal is not visible. This artefact also implies that the applied scrubbing technique is not effective against low ppm acid levels. However, adding a small amount of C_2H_4 and C_3H_6 enhances the NO_x removal efficiency.

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