Limitations of NOX removal by pulsed corona reactors

Citation for published version (APA):

Document status and date:
Published: 01/01/2011

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher’s website.
• The final author version and the galley proof are versions of the publication after peer review.
• The final published version features the final layout of the paper including the volume, issue and page numbers.

Link to publication

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
• You may not further distribute the material or use it for any profit-making activity or commercial gain
• You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the “Taverne” license above, please follow below link for the End User Agreement:
www.tue.nl/taverne

Take down policy
If you believe that this document breaches copyright please contact us at:
openaccess@tue.nl
providing details and we will investigate your claim.
The Fourth Central European Symposium on Plasma Chemistry

August 21-25, 2011, Zlatibor, Serbia

Book of Abstracts
Eds. M. M. Kuraica and B. M. Obradović

Organized by:
Faculty of Physics, University of Belgrade
Studentski Trg 12, Belgrade, Serbia

Sponsored by:
Ministry for Education and Science, Republic of Serbia, and

CENTRAL EUROPEAN INITIATIVE
LIMITATIONS OF NO\textsubscript{X} REMOVAL BY PULSED CORONA REACTORS

E.A. Filimonova\textsuperscript{1}, F.J.C.M. Beckers\textsuperscript{2}, W.F.L.M. Hoeben\textsuperscript{3}, C. Li\textsuperscript{3,4}, A.J.M. Pemen\textsuperscript{3}, E.J.M. van Heesch\textsuperscript{3}, and U. Ebert\textsuperscript{3,4}

\textsuperscript{1} Joint Insti. High Temp., Russian Acad. Sci., Izhorskaya st. 13, 2 building, 125412 Moscow, Russia
\textsuperscript{2} Oranjewoud-HMVT, P. O. Box 8590, 3009 AN Rotterdam, The Netherlands
\textsuperscript{3} Eindhoven University of Technology, P.O.Box 513, 5600 MB Eindhoven, The Netherlands
\textsuperscript{4} Centrum Wiskunde and Informatica (CWI), P.O.Box 94079, 1090 GB Amsterdam, The Netherlands

e-mails: helfil@mail.ru, e.j.m.v.heesch@tue.nl, ebert@cwi.nl

1. INTRODUCTION

One of the obstacles to use a pulsed corona discharge in industrial applications is that NO\textsubscript{x} concentration below a level of 1 ppm cannot be removed. Streamer discharges (pulsed corona discharges and dielectric barrier discharges) produce NO\textsubscript{x} themselves, and the amount of NO\textsubscript{x} depends on the deposited energy. There are presently only a few papers investigating this problem \cite{1,2}. The authors of \cite{1} suggested covering the electrodes with the photocatalyst TiO\textsubscript{2} to remove NO\textsubscript{x} concentrations below 5 ppm. The NO\textsubscript{x} removal efficiency with or without photocatalyst varied by not more than 10 \% on a total removal efficiency of 30\%.

In \cite{2}, the NO\textsubscript{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\textsubscript{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\textsubscript{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\textsubscript{x} removal can be improved by adding hydrocarbons.

2. RESULTS AND DISCUSSION

2.1. Experiments

The setup \cite{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 \cite{4}. Together with other design rules this ensures proper EMC (electro-magnetic compatibility) \cite{4}. The NO\textsubscript{x} levels are measured with Airpointer (Recordum Austria) chemoluminescence detector. We expect that the Airpointer also responds to HNO\textsubscript{3}, HNO\textsubscript{2}, N\textsubscript{2}O and N\textsubscript{2}O\textsubscript{5}. Ozone, produced by pulsed corona, was removed to below 500 ppb in a heated borosilicate glass tube (350 C) before entering the NO\textsubscript{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\textsubscript{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.

In the figure, experimental and calculation results are compared for air with 100% humidity and \([\text{NO}]_0=1 \text{ ppm}\). The agreement with experiment is better when significant components such as nitrogen oxides and nitrogen-containing acids are included. The value of \([\text{N}_2\text{O}_5]\) is almost zero. \([\text{N}_2\text{O}]\) is lower than 0.2 ppm for \(E=0.0131 \text{ J/cm}^3\). In the corona discharge, \(\text{OH}, \text{H}, \text{N}, \text{and O radicals are produced in each pulse. NO is produced mainly in the reactions } \text{O}_2 + \text{N} => \text{O} + \text{NO} \text{ and } \text{OH} + \text{N} => \text{H} + \text{NO} \text{. In humid air when } [\text{NO}] ~ 0, \text{NO}_2 \text{ is produced by the reactions } \text{OH} + \text{HNO}_2 => \text{H}_2\text{O} + \text{NO}_2, \text{ and } \text{O}_2 + \text{HNO} => \text{OH} + \text{NO}_2, \text{ and } [\text{NO}_2] \text{ decreases mainly in the reaction } \text{OH} + \text{NO}_2 + \text{M} => \text{HNO}_3 + \text{M}\). One source for acids is hydrated ions. The simulation shows that adding a small amount of \(\text{C}_2\text{H}_4\) and \(\text{C}_3\text{H}_6\) enhances the \(\text{NO}_x\) removal efficiency and decreases the concentration of acids.

CONCLUSIONS

Sub-ppm \(\text{NO}_x\) removal by pulsed corona encounters a number of serious difficulties. In the low ppm range a pulsed corona \(\text{NO}_x\) removal is balanced by a pulsed corona \(\text{NO}_x\) production. Hence, the removal process quenches below the 1 ppm \(\text{NO}_x\) level. Secondly, the chemoluminiscence measuring principle for \(\text{NO}_x\) detection also responds to nitrous oxides other than \(\text{NO}\) and \(\text{NO}_2\), and acids. Therefore, although \(\text{NO}_2\) has been converted to \(\text{HNO}_3\), the achieved \(\text{NO}_2\) 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 \(\text{C}_2\text{H}_4\) and \(\text{C}_3\text{H}_6\) enhances the \(\text{NO}_x\) removal efficiency.

ACKNOWLEDGMENTS

The authors acknowledge financial support by the Dutch Technology Foundation STW through projects 10118 and 10751, the Dutch SenterNovem-program IOP-EMVT and the companies Oranjewoud and HMVT.

REFERENCES