Proceedings of
1999 Japan-Korea Joint Symposium
on
Electrical Discharge and
High Voltage Engineering

25 - 26 October, 1999

Kyushu Institute of Technology, Kitakyushu, Japan

Organized by

Technical Committee on Electrical Discharge in IEEJ
Kyushu Institute of Technology

Co-organized by

Electrical Discharge and High Voltage Society in KIEE

In Cooperation with

The Japan Research Group of Electrical Discharges
IEEE Fukuoka Chapter

SOME ASPECTS OF CORONA DISCHARGE IN MIXTURES CONTAINING NO_X-CO_X

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The use of corona discharge is one of solutions how to remove NO_x from a flue gas. This non-thermal plasma technique has been widely used in many laboratory and pilot-scale experiments over last decade. The process of NO_x treatment, chemical reactions, final products, energy consumption and efficiency can be influenced by change of different parameters and discharge conditions. Except electrical parameters (shape of HV waveform - its amplitude, duration and frequency, discharge polarity, DC bias, etc.) the process can also be influenced by initial gas composition and different chemical additives.

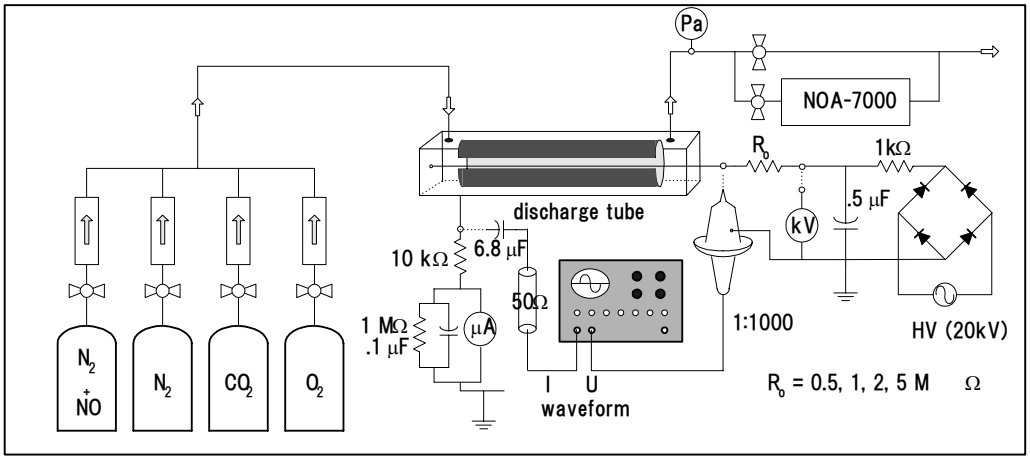
Among different additives with significant effect on deNO_x process (e.g. NH_3, H_2O, hydrocarbons, etc.) partially also an influence of CO_x had been studied. Despite of the fact that there is an assumed influence of CO_x either on a discharge behavior or NO_x treatment, so far no detailed description of the CO_x effect has not been published. According to authors [**1,2**] in this process NCO radical plays an important role. This radical can cause direct removal of either NO or NO_2 and furthermore NCO radical can also be incorporated in different heterogeneous organo-metal compounds on a surface of electrode. According to authors [**3**] such compounds are effectively created on materials like for instance copper (or brass) having significant catalytic properties.

Recently it was realized [**3**] that in some cases discharge process in mixtures CO_2-N_2 including H_2O can lead moreover to formation of aminoacids on the surface of electrodes. Here again responsible agent for the formation of aminoacids seems to be NCO radical. Its reaction with the H_2O (presence of H_2O is necessary for the process) in mixture leads to origin of amid group $^{+}NH_2$-COO^{-}, which is the beginning of the essential aminoacids' (e.g. glycine) formation process.

While discharge process in mixture CO_2-N_2-H_2O could cause the formation of aminoacids on electrode surface, the question is now whether similar process could not exist also in NO_x-CO_x-H_2O including mixtures, where NO_x and CO_x would be simultaneously removed. The aim of our present and future research project concerning NO_x is to perform measurements in corona discharge and investigate changes in mixtures containing NO_x and CO_x in presence of water. Our research should give the answer about products of the process (aminoacids or some other interesting compounds?) as well as the influence of CO_x presence on deNO_x efficiency, energy consumption and other parameters. So far only few basic experiments in the mixtures containing NO_x and CO_x have been done.

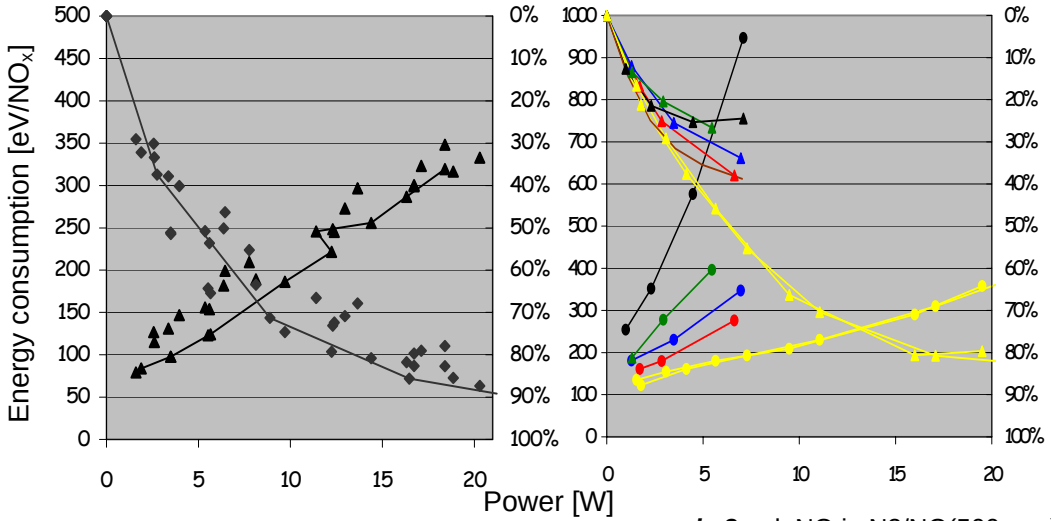
The experimental setup used in our last experiments is presented on the picture (***pic.1***). As far as at the time of measurements the qualitative analysis of discharge products in gas mixtures was absent (only chemiluminescence NO_x analyzer was available), only quantitative measurements of NO_x treatment have been performed. The experimental setup consisted of hemi-cylindrical corona reactor (copper wire ∅0.2mm / copper hemi-cylinder ∅35mm) powered by DC high voltage power supply. As

a gas mixture N_2/NO respectively N_2/NO/CO_2 was used. The gas flow of the mixture was set to 2 l/min and deNO_x efficiency of a discharge action was analyzed by chemiluminescence NO_x analyzer.

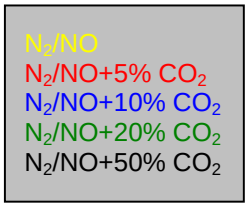


pic.1.

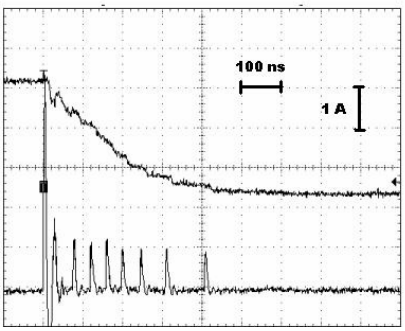
In the experiment DC corona discharge streamer mode was used exclusively. The effect of streamer corona discharge on gas mixtures and efficiency of deNO_x process in presented on the pictures (***pic.2, pic.3***) as well as the typical discharge waveform (***pic.4***)



pic.2. : deNO in N2/NO(500ppm) mixture



pic.3. : deNO in N2/NO(500ppm)/CO2(%) mixture



As it can be seen from pictures (**pic.2.**) maximum deNO treatment efficiency was more than 80% (520→100ppm) while at the same time energy consumption was 256eV/NO. Simultaneously total deNO_x treatment efficiency went on 70% (520 → 155 ppm) with 287eV/NO_x energy consumption. In the case of CO_2 presence in the mixture (**pic.3.**) deNO/deNO_x efficiency decreased, since reduction also oxidation processes took place here and so deNO_x efficiency got worse. However it is expected that in the presence of H_2O in the mixture NO_x-CO_2 the

pic.4. : typical discharge waveform (U = 8 kV, I_{mean} = 1.8 mA, f = 20 kHz)

deNO$_x$ efficiency would have get better since OH radicals play important role in NO_2 oxidation process. Presence of H_2O is necessary also for eligible formation of aminoacids **[3]**. According to authors in the case of aminoacids concentration of water should be at least as big as the concentration of CO_2 or even more. That is why we want our further experiments direct this way and use water from ultrasound humidifier in a gas mixture. However also change in apparatus is necessary.

It appeared that cylindrical geometry is very hard to control and for basic research is much better to use rather more simple geometry (e.g. point-to-plane). That is why in future we want to concentrate especially on measurements in such geometry while using simple DC corona discharge. The aim is to look after discharge mode influence on a certain gas mixture and the final products of the discharge process. There are many parameters to be varied (shape of point electrode, discharge gap, serious resistance, discharge polarity, gas mixture and gas flow) and so many experiments has to be done. First we want to concentrate on N_2-CO_2-H_2O mixtures and further on NO_x-CO_x-H_2O and compare the results and evaluate influence of CO_2 and H_2O on deNO$_x$ treatment and chemical processes in discharge volume. Analysis of gas mixture in the volume will be performed by FT-IR. The research should also include the analysis of solid structures created the surface of electrodes by heterogeneous reactions and confirm eventual presence of aminoacids or other interesting compounds.

[1] Arquilla M et al. (1993) "*Reduction of NO_x in Medium Pressure Process Flue Gas*", Int. Symp. Plasma Chem. (Proceedings), Loughborough (UK), p.593

[2] Morvová M (1994) : "*The Effect of DC Corona Discharge with Various Electrode Materials on Exhaust Components*", 10th Symp. on Elem. Proc. & Chem. Reac. in Low Temp. Plasma (Proceedings), Stará Lesná (SK), pp.155-183

[3] Morvová M, Hanic F, Morva I (1998) : "*Chemical and Physical Processes in Conversion of Gaseous System CO_2-N_2-H_2O into a Solid Condensate of Amino Acids*", 11th Symp. on Elem. Proc. & Chem. Reac. in Low Temp. Plasma (Book of Invited papers), Liptovský Ján (SK), pp.237-241