Effect of catalysts location on performance of DC corona discharge

S. Pekárek

Czech Technical University in Prague, FEE, Technická 2, 166 27 Prague 6, CR

The processes of ozone and nitrogen oxides production by the DC corona discharge can be affected not only by presence of catalysts but also by their location in the discharge chamber. For the DC hollow needle to mesh corona discharge enhanced by the flow of air through the needle electrode we studied the effect of location of TiO_2 or ZMS-5 catalysts on the mesh electrode on ozone and nitrogen monoxide/dioxide production. We found that maximum ozone concentration is obtained when TiO_2 is placed in the central region of the mesh electrode; placement of TiO_2 or ZMS-5 on the mesh decreases energy density of the onset of the nitrogen monoxide production and production of NO_2 is strongly influenced by the position of any of these catalysts on the mesh electrode.

1. Introduction

Non-thermal electrical discharges in air at atmospheric pressure are efficient source of ozone, nitrogen oxides and other reactive species. The main ozone generating reaction dominant at atmospheric pressure is

$$O + O_2 + M \rightarrow O_3^* + M \rightarrow O_3 + M,$$

where M is a third-body collision partner, which in air is O_2, N_2, or a solid metallic/dielectric surface [1]. In the case of ozone generation from air, the existence of nitrogen molecules causes the appearance of various types of positive nitrogen ions, nitrogen atoms and excited atomic and other molecular species which on the one hand produce additional oxygen atoms for ozone generation but, on the other hand, cause discharge poisoning.

The basic idea behind our research was to affect the DC needle-to-mesh corona discharge ozone and nitrogen oxides production by placing various catalysts in different regions on the mesh electrode and to find the correlation among the concentration of ozone, nitrogen monoxide and nitrogen dioxide. For our experiments we tested two catalysts. The first one was titanium dioxide TiO_2 and the second one was ZMS-5 zeolite.

The TiO_2 can be activated by the UV radiation emitted by the discharge. The zeolites such as ZMS-5 and certain ion exchanged ZSM-5 zeolites are tested for NO decomposition.

A zeolite is an inorganic porous material having a highly regular structure of pores and chambers that allows some molecules to pass through, and causes others to be either excluded or broken down. For their activation can be in some cases used the increased temperature [2,3] and in some cases the UV radiation even at temperatures as low as 275 K [4,5,6]. The objective of this paper therefore was to investigate the effect of location of TiO_2 or ZMS-5 catalysts on the mesh electrode on ozone and nitrogen monoxide/dioxide production of the DC hollow needle to mesh corona discharge enhanced by the flow of air through the needle electrode.

2. Experimental arrangement

The experimental arrangement is shown in Fig. 1. The electrodes - stainless steel hollow needle and a stainless steel mesh situated perpendicularly to the needle - were situated in a circular glass discharge chamber. The needle had an inner and outer diameter 0.7 mm and 1.2 mm respectively. The mesh had rhombus cells dimensions 0.60×0.50 mm.

Air from a cylinder was through the moisture trap and a mass flow controller MFC supplied to the needle. We also measured relative humidity RH of input air and temperature T of the output air.

The Technix DC regulated high voltage power supply provided voltage up to 30 kV. The needle, biased negatively, was ballasted by a resistor R_B.

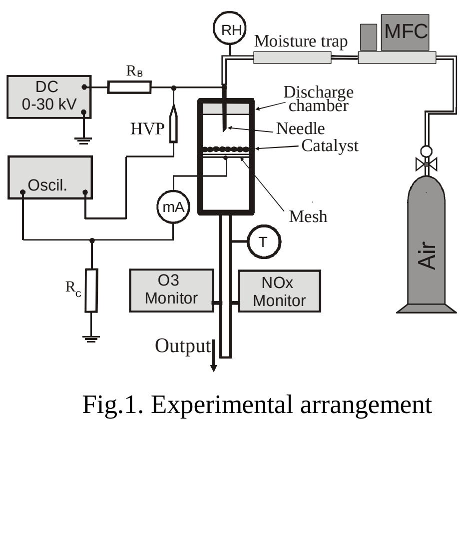

Fig.1. Experimental arrangement

The ozone concentration was measured by absorption of the 254 nm UV spectral line using an API 450 ozone monitor. Concentration of nitrogen monoxide and dioxide was measured by toxic gases monitor GA-60 equipped with 5 electrochemical cells to measure concentrations of nitrogen monoxide, nitrogen dioxide, oxygen, carbon dioxide and sulphur dioxide. Calibration of the monitor on nitrogen monoxide and nitrogen dioxide was performed using different mixtures of nitrogen monoxide and dioxide in synthetic air.

To investigate the effect of location of the catalyst on ozone and nitrogen oxides discharge production we placed on the mesh a cylindrical Teflon divider. This element divided the area of the mesh into central region and four peripheral regions. The dimensions of the central and peripheral regions of the divider were chosen taking into account two aspects. First of all when the catalyst was placed on the mesh in the central region it was in direct contact with the discharge. When the catalyst was placed on the mesh only in the peripheral regions it was not (for a distance *d*=8.1 mm between the tip of the needle and the mesh) in contact with the discharge. Secondly the dimensions of the regions were chosen in such a way that it was possible to place either into the central or into the peripheral region the same mass of the reference catalyst.

The experiments were performed with two catalysts. The first one was titanium dioxide TiO_2 Aerolyst 7706 photocatalyst. The cylindrical globules were ~3 mm in diameter and ~4 mm in height. The mass of this catalyst was m=0.7691 g. The second catalyst tested was ZSM-5 zeolite in the form of irregular spheres of whitish color ranging in diameter between 1–2 mm. When this catalyst was placed on the mesh in the central region in such a way that it filled all its area its mass was m=0.52026 g. On the other hand when this catalyst was placed on the mesh in all peripheral regions its mass was m=0.68021 g (mass of the catalyst in the first peripheral region was m_1=0.1698 g; mass of the catalyst in the second peripheral region m_2=0.17012 g; mass of the catalyst in the third peripheral region m_3= 0.1721 g and mass of the catalyst in the fourth peripheral region was m_4= 0.17013 g).

Electric current was determined as the mean value of the signal obtained from the voltage drop on the earthed resistor R_C. This signal was recorded on the first channel of the ADS 1102 CM digital oscilloscope (bandwidth 100 MHz). The discharge voltage U was determined as the mean value of the signal recorded on the second channel of this oscilloscope through the high voltage probe HVP.

3. Experimental results

All experiments without as well as with the catalyst were performed with the divider on the mesh. The airflow through the needle was 1.8 slm and the relative humidity of air 0.8 %. The needle was biased negatively. The distance between the tip of the needle and the mesh was 8.1 mm. The results presented in following figures are average values obtained from five measurements.

It should be pointed out that placement of various catalysts in different regions on the mesh changes not only electrical parameters of the discharge but at the same time also production of ozone as well as nitrogen oxides. This is why these quantities could not be considered separately but one in the context with others. Thus the graphs labeled as a) in Figs. 2 - 6 show the volt-ampere characteristics of the discharge. The graphs labeled as b) in these figures show the dependences of ozone concentration on the mean discharge current and the onset of the discharge poisoning. The graphs labeled as c) show the dependences of nitrogen monoxide concentration (for Figure 6 for the needle to mesh electrodes with ZSM-5 in the central region also the nitrogen dioxide) and finally the graphs labeled as d) the dependences of nitrogen dioxide concentration versus mean discharge current.

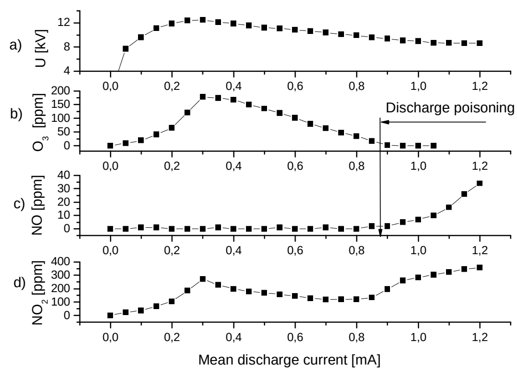

Fig.2. Dependence of the a) discharge voltage; b) ozone concentration; c) concentration of nitrogen monoxide and d) concentration of nitrogen dioxide on the mean discharge current for the discharge without any catalyst.

From Figs. 2–6 can be seen that the maximum discharge ozone production takes place when the discharge voltage reaches its maximum. It is also seen that placement of either TiO_2 or ZMS-5 in the central region of the mesh increases current at which maximum ozone production occurs. Placement any of these two catalysts in the peripheral region of the mesh does not substantially affect the value of this current.

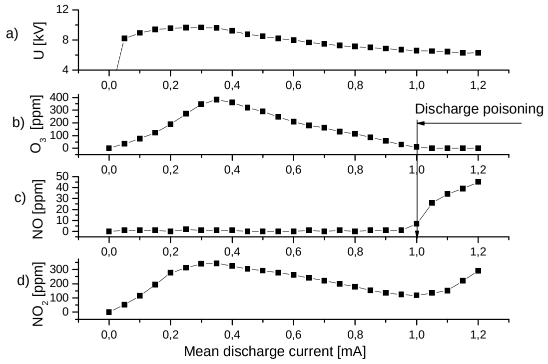

Fig.3. Dependence of the a) discharge voltage; b) ozone concentration; c) concentration of nitrogen monoxide and d) concentration of nitrogen dioxide on the mean discharge current for the discharge with TiO_2 in the central region. m_{TiO2}= 0.7691 g.

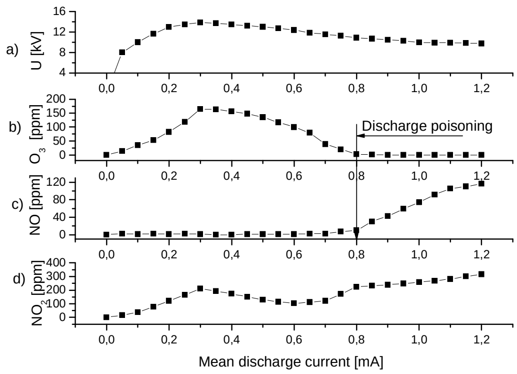

Fig.4. Dependence of the a) discharge voltage; b) ozone concentration; c) concentration of nitrogen monoxide and d) concentration of nitrogen dioxide on the mean discharge current for the discharge with TiO_2 in the peripheral regions, central region free. m_{TiO2}= 0.7691 g.

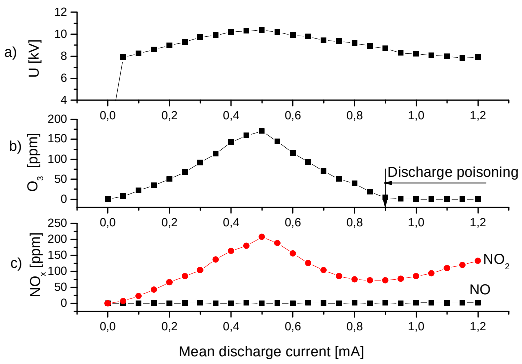

Fig.5. Dependence of the a) discharge voltage; b) ozone concentration; c) concentration of nitrogen monoxide and nitrogen dioxide on the mean discharge current for the needle to mesh electrodes with ZSM-5 in the central region. m_{ZSM-5}= 0.52026g.

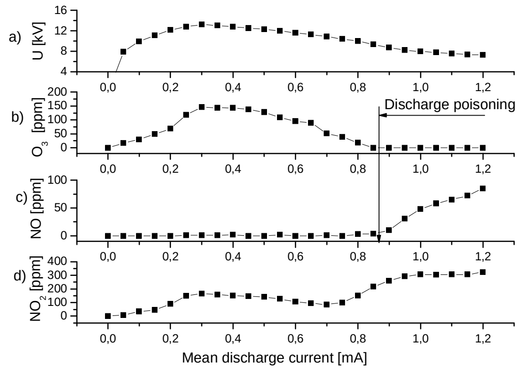

Fig.6. Dependence of the a) discharge voltage; b) ozone concentration; c) concentration of nitrogen monoxide and d) concentration of nitrogen dioxide on the mean discharge current for the discharge with ZSM-5 in the peripheral regions, central region free. m_{ZSM-5}= 0.68021 g.

From Figs. 2–6 can be also concluded that for the discharge with a simple mesh as well as for the discharge with any of these two catalysts in the central as well as in the peripheral regions the maximum discharge voltage, maximum concentration of the produced ozone and maximum concentration of the produced nitrogen dioxide occur for the same currents. It is also seen that for the discharge with a simple mesh electrode as well as for the discharge with each of the catalysts at the central as well as at the peripheral regions the value of current of the onset of the discharge poisoning coincides with the current when the discharge starts to produce the nitrogen monoxide.

From the comparison of V-A characteristics of the discharge with simple mesh electrode and with TiO_2 or ZMS-5 in the peripheral region can be concluded that placement of any of these catalysts in the peripheral region on the mesh increases discharge voltage (in case of ZMS-5 for current smaller than 0.8 mA) in comparison with a discharge with a simple mesh only.

To compare the results dealing with ozone and nitrogen oxides production for the discharge with a simple mesh and for the discharge with catalysts in different regions on the mesh it is necessary to take into account that the placement of catalysts different regions on the mesh changes in a different way electrical parameters of the discharge. To involve this effect into the analysis we present the concentrations of ozone and nitrogen oxides not as a function of the mean discharge current but as a function of energy density, which is defined as a ratio of the mean power delivered to the discharge and the flow of air through the needle electrode.

The dependence of ozone concentration versus energy density for the discharge with a needle to mesh electrode and a mesh electrode with a layer of TiO_2 or ZSM-5 in central or in peripheral regions is shown in Fig. 7. Similar dependence however for the nitrogen monoxide is shown in Fig.8. Both these figures are for the mass of TiO_2 globules m=0.7691 g, mass of ZSM-5 in the central region m=0.52026 g and mass of ZSM-5 in all peripheral regions m=0.68021 g.

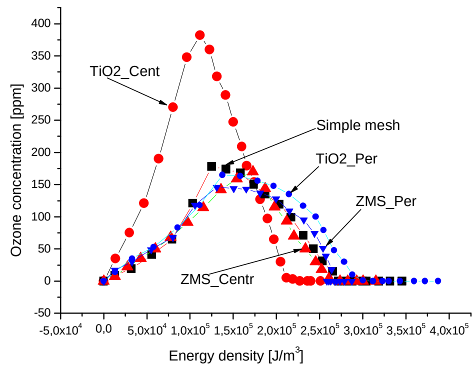

Fig.7. Ozone concentration versus energy density for the discharge with the needle to mesh electrodes; with the needle to mesh electrodes with TiO_2 or ZSM-5 in the central region or in the peripheral region.

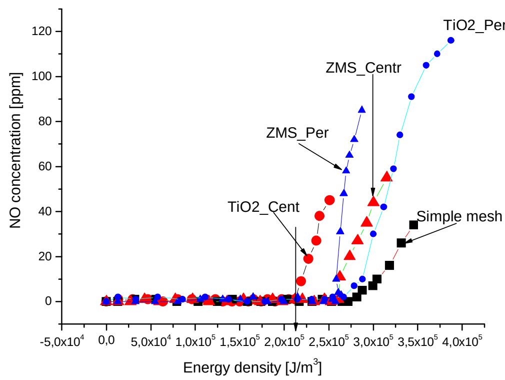

Fig.8. Nitrogen monoxide concentration versus energy density for a) needle to mesh electrodes; b) needle to mesh electrodes with TiO_2 in the central region; c) needle to mesh electrodes with TiO_2 in the peripheral region; c) needle to mesh electrodes with ZMS-5 in the central region and d) needle to mesh electrodes with ZMS-5 in the peripheral regions.

The differences in discharge ozone and nitrogen oxides production when TiO_2 or ZMS-5 catalysts are placed in different regions on the mesh electrode can be explained mainly by:

- Different electrical properties of these catalysts.
- Different mechanism of their activation.

4. Conclusions

We have investigated the effect of location of titanium dioxide TiO_2 or ZMS-5 zeolite in the discharge region on the production of ozone, nitrogen monoxide and nitrogen dioxide for a DC hollow needle to mesh corona discharge enhanced by the flow of air through the needle electrode. Our findings can be summarized as follows:

- Maximum ozone concentration is obtained when TiO_2 is placed in the central region of the mesh electrode.
- Placement of TiO_2 or ZMS-5 on the mesh electrode decreases energy density of the onset of the nitrogen monoxide production.
- Production of NO_2 is strongly influenced by the position of any of these catalysts on the mesh electrode.

5. Acknowledgement

This research has been supported by the Czech Science Foundation grant under contract No. 202/09/0176.

6. References

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