NEGATIVE CORONA DISCHARGE IN SYNTHETIC AND DRY AIR

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Electrical discharges in air generate many different species, some of them toxic and corrosive. Ozone is a predominant species, but also nitrogen oxides, such as N_2O, NO, NO_2, NO_3 and N_2O_5, have been confirmed by means of spectroscopic techniques and mass spectrometry in air-fed ozonizers based on dielectric barrier discharge [1,2]. The production of chemical species by means of corona discharge in air has received less attention [3,4] and, although the fundamental physico-chemical processes in dielectric barrier and corona discharges are essentially identical, both discharges are very different from the electrical point of view.

In this work, a numerical modelling of negative DC corona discharge in dry air (0.03% CO_2) is performed for wire-to-cylinder coaxial geometry. The chemical kinetic model used here includes nine charged species (e, N_2^+, NO^+, NO^-, O_2^+, O_2^-, O^-, O_3^- and CO_3^-), ten neutral species in their ground states (N_2, O_2, ,CO_2, N, NO, N_2O, NO_2, O, O_3 and CO), four excited species ($N_2(A^3\Sigma_u^+)$, $N(^2D)$, $O_2(^1\Delta_g)$ and $O(^1D)$), with a total number of 150 reactions. The electrical discharge is assumed to be homogeneous along the wire and stationary in time. Therefore, the concentration of species will only depend on the radial distance to the wire (1D model). The equations governing the electrical discharged are continuity equations for each species (which include the chemical kinetics) and Poisson's equation for the electric field. The electric field dependence of reaction rate constants has also been taken into consideration, particularly in electron induced reactions.

The numerical modelling is complemented with the experimental measurements of ozone and nitrogen dioxide concentrations generated by a negative corona discharge in synthetic air. The electrode arrangement used in the experiments (tungsten wire $\varnothing$ 0.1 mm, stainless steel
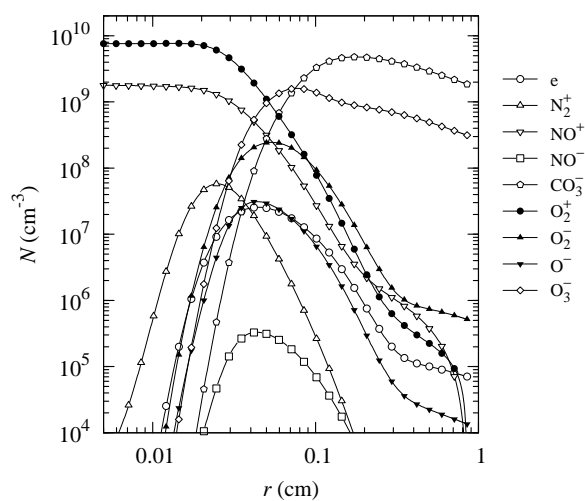

Fig. 1 Radial distribution of electrons and ions corresponding to an applied voltage of -6.5 kV (numerical simulation).
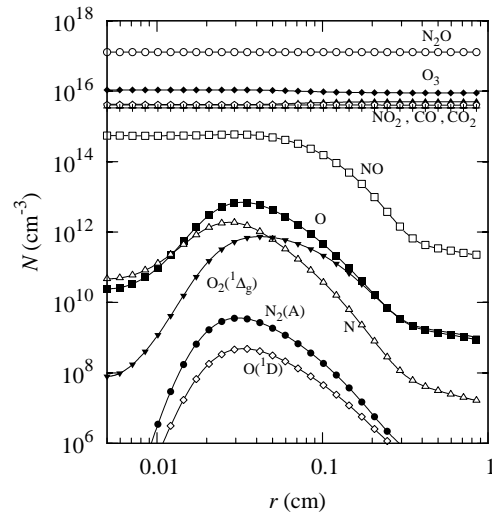

Fig. 2 Radial distribution of neutral species corresponding to an applied voltage of -6.5 kV (numerical simulation).
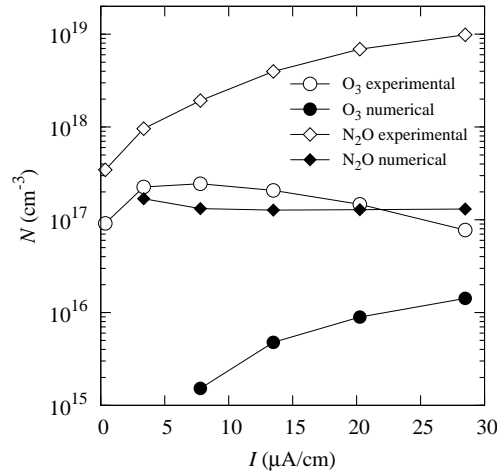
Fig. 3 Averaged ozone and nitrous oxide densities as a function of corona current intensity

cylinder $\varnothing$ 17 mm) is identical to the one considered in the numerical simulation. The gas inside the corona discharge reactor was prepared as a mixture of 20% oxygen and 80% nitrogen, and species concentrations were determined by means of UV absorption spectroscopy, in the range 190-330 nm. The high voltage was applied from 4.5 kV (slightly above the onset of corona) up to 7.5 kV. Each applied voltage was maintained for about 50 minutes so as to enable that a stationary current and, presumably, stationary species concentrations, were reached.

Figures 1 and 2 show the radial distribution of charged and neutral species, respectively, according to the numerical simulation for an applied voltage of -6.5 kV and a current intensity of -20.3μA/cm. In the vicinity of the wire, ionization of nitrogen and oxygen molecules by electron impact gives rise to the formation of N_2^+ and O_2^+. However, O_2^+ ions are predominant due to the existence of a fast charge transfer reaction from N_2^+ to oxygen molecules to form O_2^+. In the drift region, the principal negative ion is CO_3^-, followed by O_3^-. This result is in agreement with mass spectrometric study of ions produced by negative corona in air [5], and it can be explained by the presence of a fast ion conversion reaction between O_3^- and CO_3^-. Concerning the radial distribution of neutral species (Fig. 2), the predominant species is N_2O, which is formed at the expenses of NO_2 and atomic nitrogen. Moreover, ozone density is almost two orders of magnitude smaller than in the case of corona in pure oxygen [6], even though the density of oxygen has only been reduced by a factor of five. This dramatic reduction of ozone is the consequence of catalytic reactions chains, which lead to the recombination of oxygen atoms. In figure 3, the averaged ozone and nitrous oxide densities measured in the experiments are compared with the results obtained from the numerical simulation at different current intensities. As predicted by the numerical simulation, the concentration of N_2O was always higher than that of O_3, but the numerical simulation tends to underestimate the actual concentrations.

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