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Experimental study of positive corona discharge in mixtures of CO_2 and N_2

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Positive corona discharge in mixtures of CO_2 and N_2 has been experimentally investigated using a coaxial wire-to-cylinder corona discharge reactor. Special attention has been paid to the stability of the corona current and to the generation of ozone and nitrogen oxides in time, which are influenced by the applied voltage and the ratio of CO_2:N_2 in the gas mixture.

Carbon dioxide is a green house gas with an important contribution to the global warming of the atmosphere. The decomposition of carbon dioxide through the reactions induced by electrical discharges has long been considered as a promising possibility in CO_2 abatement technologies. Therefore, many past studies have paid attention to the decomposition of CO_2 under the effect of DC and pulsed corona discharge and the production of other species of industrial interest, like ozone [1-4].

In this paper, we have conducted an experimental research of positive DC corona discharge in mixtures of carbon dioxide and nitrogen. The corona discharge reactor used in the experiments consisted of a tungsten wire (the anode) with radius r = 0.0625 mm, situated along the axis of a stainless steel cylinder (the anode) with inner radius R = 0.85 cm. Both electrodes were 5 cm long, and the cell was closed with UV grade windows at each end of the cylinder. The discharge reactor was filled with a mixture of high purity CO_2 (99.995%) and N_2 (99.999%) and it was placed inside the sample compartment of a Thermo Nicolet UV/VIS spectrophotometer. The high voltage was applied to the corona wire for 45 minutes, and periodic measurements of the corona current (every second) and of the UV spectrum (every 300 s) were taken during that time.

The time evolution of the current intensity corresponding to V = 7 kV and 8 kV are presented in figure 1a and 1b, respectively, for different concentrations of N_2 in the gas mixture. In our experimental set-up, the onset of positive corona discharge occurs below 6.5 kV. However, for 6.5 kV and 7 kV (fig. 1a), the corona current becomes unstable after some time if the concentration of N_2 is

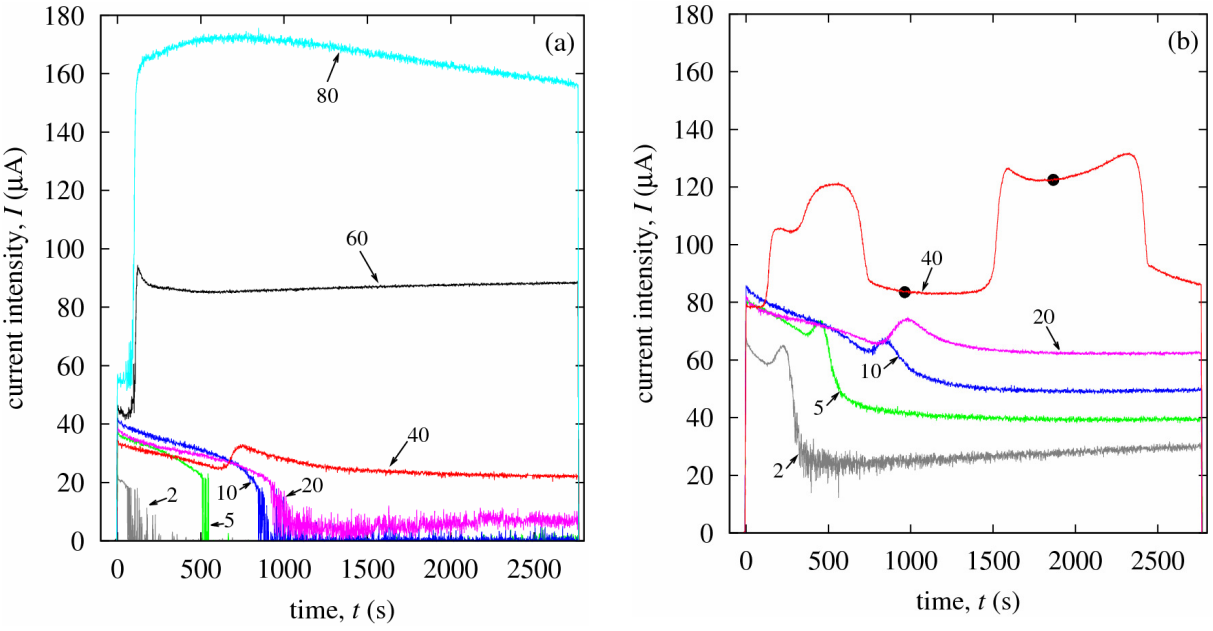

Fig. 1: Time evolution of corona current intensity in mixtures of CO_2 and N_2 for (a) V = 7 kV and (b) V = 8 kV. The volume percentage of N_2 corresponding to each line is indicated in the figure.

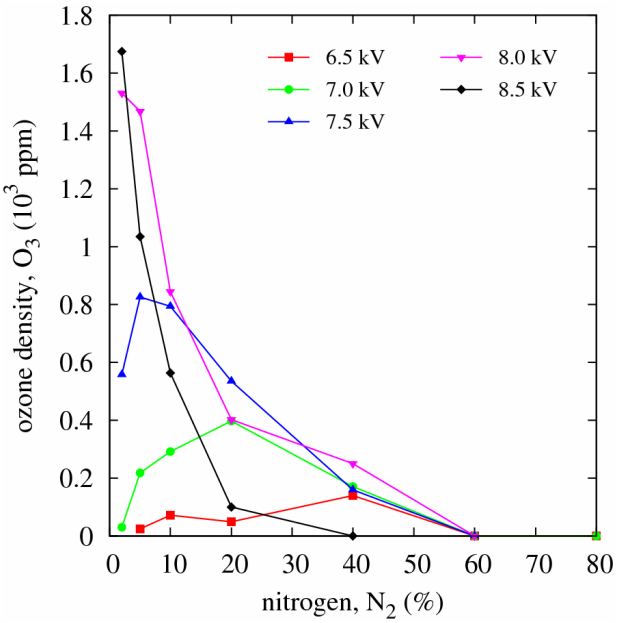

Fig. 2: Ozone density as a function of the initial nitrogen concentration for different applied voltages. The values of ozone density shown correspond to 45 minutes after the application of the voltage.

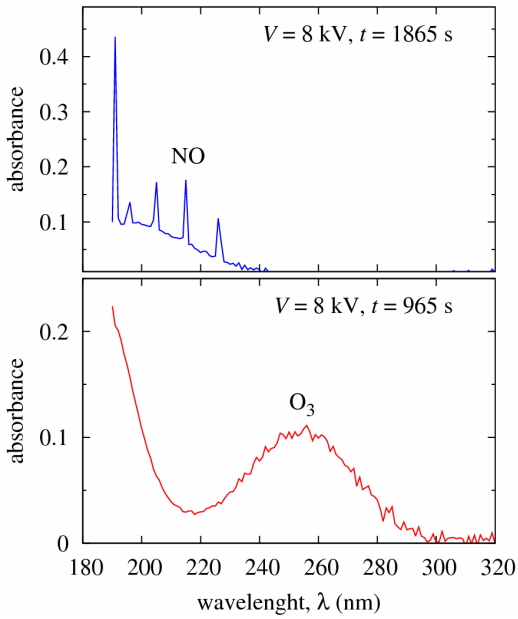

Fig. 3: UV spectra of a gas mixture of 40% N_2 and 60% CO_2) for an applied voltage V = 8 kV and at two different times: 1865 s (top) and 965 s (bottom). These times are indicated with a black dot in Fig. 1.

sufficiently small (2-10% of N_2 in fig. 1a). In such cases, the corona current falls to zero or remains as a succession of pulses of small amplitude. Increasing the percentage of N_2 in the gas mixture (e.g. 40-80% for 7kV, fig. 1a) or the applied voltage (e.g. 8 kV, fig. 1b) stabilizes the corona current. However, the stable corona still exhibits a complex behavior. On the one hand, two different regimes of corona discharge are observed during the experiments, which are characterized by different levels of the corona current. The "low current" regime occurs when the content of N_2 in the gas mixture is below 40%, approximately (20-40% in fig. 1a, and 2-30% in fig. 1b). During this regime, ozone is readily detected as a result of the chemical activity of the corona discharge. As shown in figure 2, ozone concentration decreases with increasing the percentage of nitrogen, as it is no longer measurable when the percentage of nitrogen is 60% or above. In the "high current" regime (60-80% in fig. 1a) only nitrogen oxides and, particularly, nitric oxide (NO) are detected using UV spectrophotometry. The transition between the two corona regimes, while keeping the same applied voltage, is shown in figure 1b for the case 40% of N_2. The spectra in figure 3 correspond to the times marked with dots along the corona current line in figure 1b. On the other hand, during the "low current" regime, the corona current exhibits a characteristic bump at a certain time. The occurrence of this event is delayed with increasing the percentage of nitrogen (fig. 1b). This brief augmentation on the corona current is probably linked to the chemical activity that is happening inside the reactor. Unfortunately, the interval between spectra was too large to resolve these short events.

Acknowledgements

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