

Ozone generation using negative corona discharge: numerical simulation and comparison with experiments

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Ozone generation by corona discharge is the object of a continuous and renewed interest, since this type of electrical discharge is present in a wide variety of industrial devices, such as electrostatic precipitators, air cleaners, laser printers, copiers, etc. Many experimental and numerical studies have been conducted to improve our understanding of ozone production [1-6]. However, precise simulation of corona discharge is a rather sophisticated task since many different problems concur together in the modeling: the hydrodynamics motion of the gas, the chemical kinetics of ozone (as well as many other charged and neutral species) and the physical modeling of the electrical discharge itself.

In this work, the generation of ozone from pure oxygen by negative corona discharge is investigated both experimentally and numerically. The discharge reactor used in the experiments consisted of a wire-to-cylinder coaxial electrode system, with total length $L = 5$ cm. The cylinder was made of plain steel, stainless steel or aluminium, with inner radius $R = 8.5$ mm, while the cathode wire was made of tungsten, with radius $r = 0.05$ mm. The wire was subjected to negative DC high voltage, and the cylinder was connected to ground through a digital multimeter. The discharge cell was fed with high purity oxygen (99.995%), and the flow rate was regulated using a mass flow controller. The discharge cell was placed inside the sample compartment of a UV-visible spectrophotometer and ozone concentration was determined by applying Beer-Lambert's law in the range of wavelengths 190-300 nm. A schematic diagram of the experimental setup used in the experiments is presented in Figure 1.

The corona discharge was numerically simulated using a hydrodynamics model, where continuity equations for every species are coupled with Poisson's equation through charged

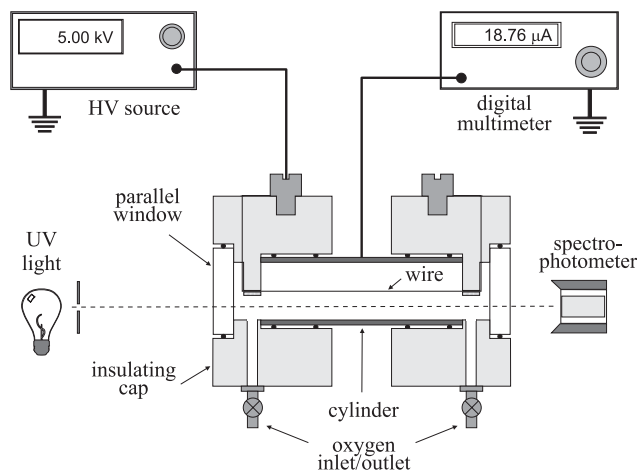


Figure 1. Schematic diagram of the experimental setup.

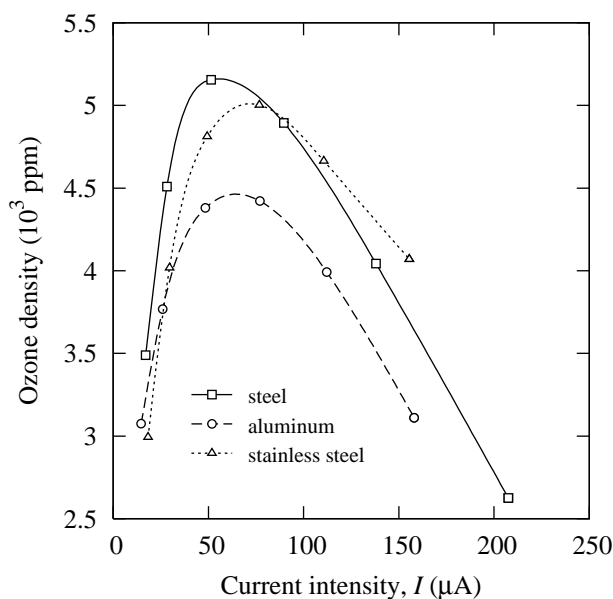


Figure 2. Averaged ozone density in the absence of gas flow as a function of the current intensity. Three different materials have been used as the anode.

species and the electric field dependence of reaction rate constants. The plasma chemical kinetics is incorporated into the model as gain/loss rate terms in the continuity equation. Finally, the effect of Joule heating was also taken into account in the energy conservation equation.

Figure 2 shows the experimental measurements of ozone concentration inside the discharge cell as a function of the current intensity and in the absence of gas flow. Ozone concentration is expressed in ppm in volume ($1 \text{ ppm} \approx 2.46 \times 10^{13} \text{ molecules/cm}^3$). The results are qualitatively similar using the three different anodes. Initially, the concentration of ozone increases sharply as the current intensity is augmented. Then, a maximum in the ozone concentration is reached, and any further augmentation of the current intensity makes the ozone concentration to decrease. The maximum ozone concentration is obtained when using the plain steel cylinder, for $I \approx 50 \mu\text{A}$.

The results obtained from the numerical simulation have shown that the observed dependence of ozone concentration with current intensity is the result of two combined effects: ozone decomposition on the electrode walls, and the existence of a significant temperature gradient inside the discharge reactor (Figure 3). If none of these effects are considered, the numerical simulation predicts an almost constant ozone concentration, independent of current intensity. However, the existence of reactions between ozone and the electrode walls constitutes an important sink of ozone molecules, particularly in the vicinity of the anode owing to its larger surface. With the inclusion of this effect, the numerical simulation predicts an increasing ozone density with current intensity, which is still unsatisfactory. This behaviour is altered when Joule heating is included into the model, since several important reactions that decompose ozone are activated by temperature. Moreover, Joule heating is more important at higher currents, which helps to limit the growth of ozone concentration with current intensity. If both effects are incorporated into the numerical model, the predictions of the simulation are in qualitative agreement with the experimental measurements, although the numerical simulation tends to overestimate the ozone concentration.

When pure oxygen is continuously fed into the reactor, the importance of the above discussed effects becomes less important, since the gas is cooled down by the presence of the gas flow and ozone decomposition on the walls is only acting during the residence time of the gas inside the cylinder. Therefore, the higher the gas flow rate is the weaker both effects will result. Figure 4 shows the experimental measurements and the predictions of the numerical simulation corresponding to two different gas flow rates. The averaged ozone concentration is expressed as a function of Becker parameter, $\beta = IV / Q$, where Q is the gas flow rate. The results of the simulation are in good qualitative agreement with the measured data: ozone concentration is

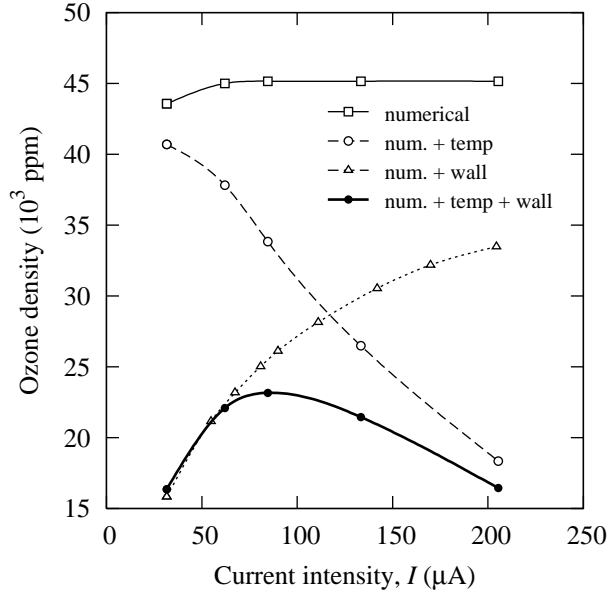


Figure 3. Averaged ozone density as a function of current intensity predicted by four numerical models of increasing complexity (num: plain model, num. + temp: including the effect of Joule heating, num. + wall: including the effect of ozone decomposition on the electrodes, num. + temp + wall: including both the effects of Joule heating and ozone decomposition on the electrodes).

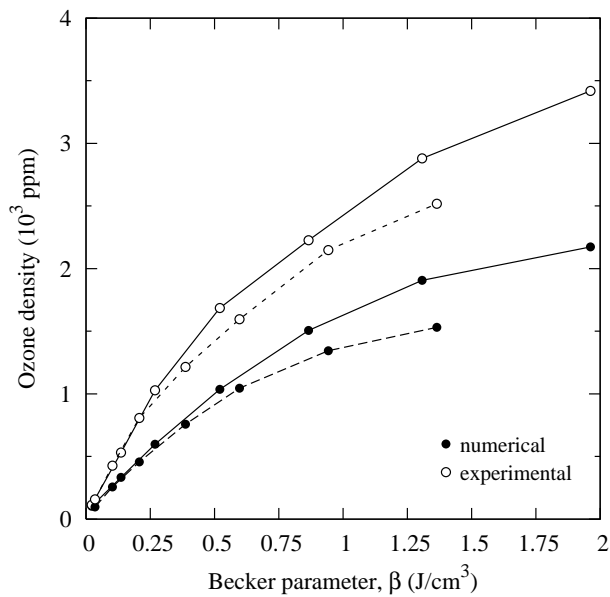


Figure 4. Experimental and numerical results of the averaged ozone density as a function of Becker parameter for two different flow rates. Solid line: $Q = 75 \text{ cm}^3/\text{min}$, dashed line: $Q = 125 \text{ cm}^3/\text{min}$.

observed to increase with Becker parameter and with a similar growth rate. Quantitatively, the numerical simulation slightly underestimates the ozone density, as the predicted ozone concentration is 35% lower than the actual measured concentration.

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