Ozone production using nanosecond-pulse and AC driven DBD in mixtures of oxygen and argon

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1. Introduction

Ozone (O_3) in a strong oxidizing agent with many industrial applications, such as bleaching, water disinfectant treatment, elimination of odors, etc [1]. It can be produced in electrical discharges and, particularly, dielectric barrier discharge (DBD) is commonly used in large installations [2]. Reducing the energy cost of ozone production is of prime importance in industrial applications and, among other factors, the voltage waveform used in stimulation of the DBD is known to play an important role [3]. Therefore, it is of interest to determine which stimulation provides the best electrical energy conversion to promote the chemical reactions that maximize the ozone yield with the highest energy efficiency.

In this work, the energy efficiency of ozone production using DBD fed with pure oxygen (O_2) and with a mixture of 50 % oxygen and 50 % argon (Ar) has been investigated. Two different forms of stimulation have been used: AC and pulses of nanosecond duration.

2. Experimental set-up and methods

Figure 1 shows a schematic representation of the experimental set-up used in the experiments. The DBD reactor consisted of two stainless steel plane circular electrodes, 20 mm in diameter, covered with fused silica glasses of 1 mm of thickness. The gap between dielectrics was 2 mm. Experiments were carried with a total gas flow rate Q = 100 cm^3/min. The concentration of ozone in the effluent gas was measured using UV/VIS absorption spectrophotometry (Thermo Evolution 300) in the wavelength interval 190 – 320 nm.

For the AC stimulated DBD, high voltage was applied to one of the electrodes using a high voltage amplifier (Trek 20/20C-HS). The voltage amplitude was set to 20 kVpp, and three different frequencies were used in the experiments: 200, 500 and 1000 Hz. The other electrode was connected to a monitor capacitor (C_m = 1 μF), and the voltage drop across the capacitor, $V_\mathrm{m}(t)$, was measured in order to calculate the power delivered to the discharge. Both the high voltage signal and the voltage drop across the capacitor were recorded using a 2.5 GHz bandwidth digital oscilloscope (Tektronix DPO7254).

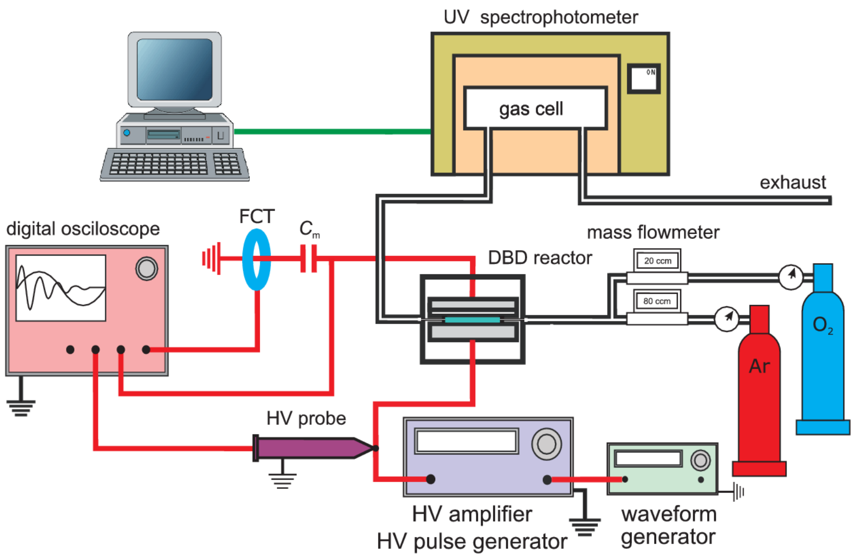

Figure 1: *Schematic diagram of the experimental setup used in AC and pulsed DBD.*

For the pulsed DBD, a high voltage nanosecond pulse generator (Megaimpulse NPG18-3500N), based on drift step recovery diodes (DSRD), was used as power supply. This generator produces high voltage pulses with rise times of less than 4 ns, and duration of about 20 ns. The rate of repetition of pulses was controlled externally with an arbitrary waveform generator (33521A, Agilent), which supplied the trigger signal to the pulse generator. Three frequencies were used in the experiments: 100, 200 and 500 Hz. The amplitude of the high voltage pulse was –21.6 kV, approximately. However, due to the highly nonlinear nature of the barrier discharge, some reflection of energy inevitably occurs at the reactor. Thus, the first pulse is followed by a subsequent train of pulses of lower amplitude. The voltage signal provided by the pulse generator was measured with a wide-band high voltage probe with 1000X attenuation (Tektronix P6015A). A fast current transformer (FCT, Bergoz), with a sensitivity of 0.5 V/A, was used for the measurement of the current intensity. Both signals were recorded using the digital oscilloscope.

In order to determine the energy efficiency of ozone production, the averaged electric power, P, and the electrical energy density, P/Q (J/cm^3), injected in the reactor must be evaluated. In the case of using AC stimulated DBD, the resulting averaged power can be obtained as

$$P = f C_{\mathrm{m}} \int_0^T V(t)\, dV_m(t)$$

where $T = f^{-1}$ is the period of the AC signal, and $V(t)$ is the voltage drop across the DBD reactor.

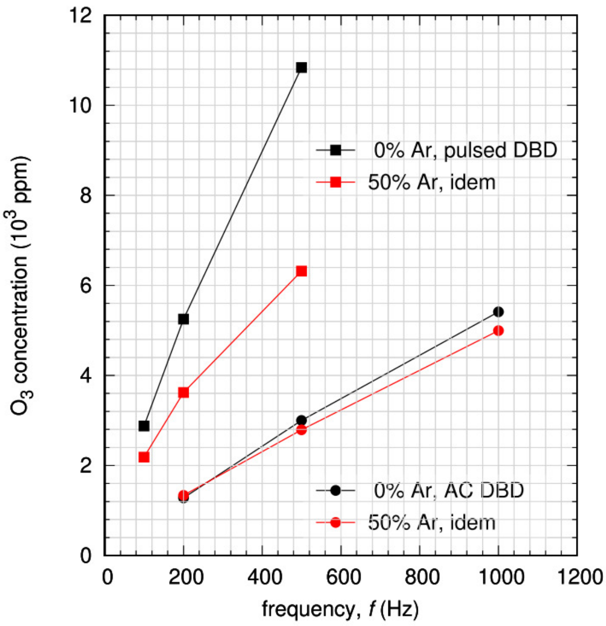

Figure 3: *Ozone yield as a function of the frequency in pure oxygen and in a mixture with argon (50 %) using AC and nanosecond pulse stimulated DBD.*

In pulsed DBD, the average power P delivered to the gas by a train of pulses (*i.e.*, the first high voltage pulse and the subsequent pulses of lower amplitude) was computed directly from the signals recorded by the oscilloscope as

$$P = f \int_0^{\tau} \bar{V}(t)\, \bar{I}(t - t_0)\, dt$$

where $\bar{V}(t)$ is the averaged voltage across the DBD reactor measured by the high voltage probe, $\bar{I}(t)$ is the averaged current intensity obtained with the FCT, and t_0 accounts for the delay between both signals due to the different lengths and properties of cables transmitting the signals to the scope. The values $\bar{V}(t)$ and $\bar{I}(t)$ were obtained by averaging over a large number of recorded samples (≥ 100), in order to reduce random fluctuations. Time integration was carried out over a sufficiently long interval, $\tau \approx 2\ \mu$s, so that the voltage amplitude of subsequent pulses becomes negligible at the end of that interval.

3. Results and discussion

Figure 2 shows the ozone concentration in pure oxygen and in a mixture of oxygen and argon (50 %) at the different frequencies investigated in this study. Clearly, the yield of ozone using pulsed DBD is greater than that obtained using AC DBD at the same frequency. Moreover, halving oxygen concentration in the gas mixture affects very differently ozone production. While in AC operated DBD the reduction of oxygen has very little or no effect,

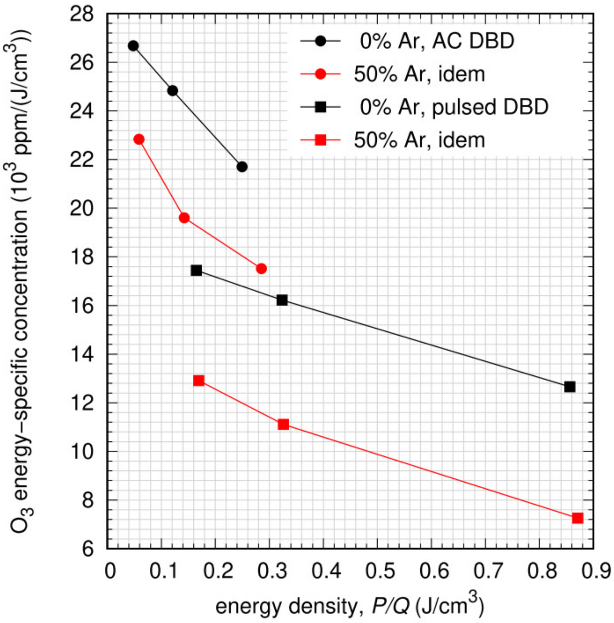

Figure 3: *Ozone energy-specific yield as a function of the frequency in pure oxygen and in a mixture with argon (50 %) using AC and nanosecond pulse stimulated DBD.*

ozone production using nanopulse driven DBD is significantly reduced, particularly at highest frequencies.

The higher ozone yield obtained by using nanopulse stimulated DBD is at the cost of lower energy efficiency. This fact can be appreciated in figure 3, where ozone concentration per unit of energy density is plotted for both AC and nanosecond pulse driven DBD. In both cases, the ozone production efficiency decreases as the operation frequency is raised. However, it must be remarked that the reduction of ozone production efficiency is faster when using AC stimulated DBD. Moreover, ozone production efficiency is always higher in pure oxygen than in the mixture of oxygen and argon, and the difference between the two efficiencies is similar in both DBDs, either driven by AC or nanosecond pulses.

Acknowledgements

This work has been supported by the Spanish Government Agency “Ministerio de Ciencia, Innovación y Universidades” and the European Regional Development Fund, under contract PGC2018-099217-B-I00.

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