Processing of COx Molecules in CO2/O2 Gas Mixture by Dielectric Barrier Discharge: Understanding the Effect of Internal Parameters of the Discharge

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Processing of CO_x molecules in CO_2/O_2 gas mixture by dielectric barrier discharge:

Understanding the effect of internal parameters of the discharge

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Abstract. Environmental pollution has become a major issue due to the rapid growth of industrial and technological developments that requires a high consumption of fossil energy. A new route of treatment of pollutant molecules bases on the use of non-equilibrium thermodynamic reactive plasmas generated by electrical discharges at atmospheric pressure to neutralize or transform toxic oxides as CO_2[1-6]. This type of non-equilibrium reactive plasma can be used for the decontamination of gaseous effluents and is generally generated by a pulsed discharge which constitutes a chemically very active medium of low energy consumption. Our work will be based on a zero-dimensional model, to study the reduction of COx in the CO_2 / O_2 gas mixture by dielectric barrier discharge of non-equilibrium plasma under typical operating conditions of the discharge. A model allows to calculate the temporal evolutions of chemical characteristics. The influence of certain discharge parameters such as the applied electric voltage, the gas pressure, the capacity of the dielectric, the discharge frequency and the concentration of oxygen in the gaseous mixture on the density variations of CO and CO_2 compared to the initial density of CO_2 in the gas mixture of the discharge have been analyzed.

Keywords: CO_2 /O_2 gas mixture, COx, pulsed DBD discharge.

1 Introduction

Mitigating greenhouse gas emissions represents an important problem in today's world. While still being the main propellant for the industrial progress, burning fossil fuels gives an ever-increasing rate of COx emission in the atmosphere. As a result, finding technological solutions for COx reduction became a rapidly growing research topic in many scientific fields. Plasmas are increasingly being used for gas conversion in both research and industrial applications [7-11], such as the destruction of large hydrocarbons, volatile organic compounds (VOCs).The constantly reducing usability of fossil fuels, in combination with the need to decrease greenhouse gas emissions, has given rise to the necessity of developing sustainable energy sources through greenhouse gas as raw materials [12-17].

2 Description of the discharge model

The electrical circuit used to deposit energy in the plasma is shown below in Fig. 1, the dielectric layers are represented by two capacitances connected in series and Cd is their equivalent capacitance.

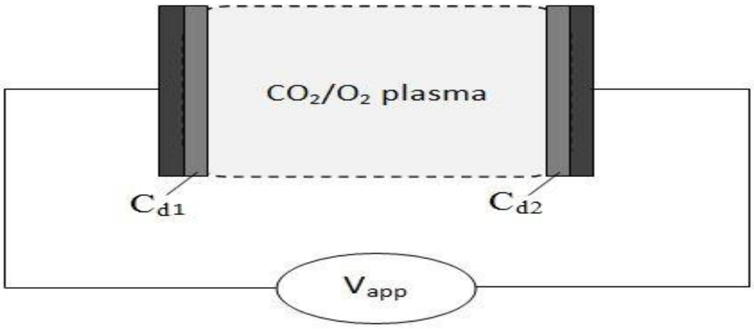

Fig. 1. Discharge model scheme used in the present calculations.

The applied voltage through the discharge is given by the following formula:

$$Vapp(t) = Vg(t) + Vd(t) \quad (1)$$

Where Vg (t) and Vd(t) are the plasma and the dielectric voltages, respectively. The voltage across the dielectrics is given by the relation:

$$V_d(t) = \frac{1}{C_d} \int I(t) dt \quad (2)$$

Where Cd is the dielectric capacitance.
The relation between the current I and voltage Vg across the gap is:

$$I(t) = \frac{V_g(t)}{R_g(t)} \quad (3)$$

The time dependence of the gap resistor Rg(t) of plasma is obtained by the following relation:

$$R_g(t) = \frac{d}{A.e.n_e(t).\mu_e(t)} \quad (4)$$

Where e, $n_e(t)$and $\mu_e(t)$are the electron charge, the time dependent electron density and mobility, respectively. *A* represents the discharge cross section in the plane of the electrodes and *d* is the separation between the discharge electrodes.

The system of equations describing the electrical circuit and the plasma kinetic is solved as follows: for a given time t, the plasma kinetic equations coupled with the electric circuit equations are solved with the classical GEAR method [18], between the instants t and t+dt.

In order to describe the electrical and chemical properties of the CO_2/O_2 plasma mixture, we have established a full set of processes involving twenty-one (21) species: e,O, O_2, O_3, C, CO, CO_2, C_2O ,O+, O_2^+, CO+, CO_2^+, O− , O_2^-, O_3^-, CO_3^- , CO_4^-,O(1D),O_2(a), O_2(b) , CO_2^* regrouped in 113chemical reactions. The rate coefficients of electron–molecule collisions, depending on the reduced electric field (E/N), are tabulated by solving the homogenous electron Boltzmann equation, and it can be obtained by the equation:

$$K_i=\sqrt{\frac{2e}{m}}\int_0^{\infty} \sigma_i \, \varepsilon f(\varepsilon) d\varepsilon \tag{5}$$

Where the parameters e, m, ε, and σi are the electron charge, the electron mass, electron energy and electron impact cross section of the process i, respectively, and f (ε) is the electron energy distribution function (EEDF).

3 The effect of internal parameters

In this subsection, we examine the effect of some discharge parameters, which are indicated below, on the CO concentration in the discharge and on the CO_2 conversion factor. The CO_2 conversion is defined as follows:

$$C_{CO_2}(\%)=\frac{[CO_2]_0-[CO_2](t)}{[CO_2]_0} \times 100 \tag{6}$$

Where $[CO_2]_0$is the initial concentration of CO_2 in the gas mixture (without plasma) and$[CO_2](t)$ is the concentration of CO_2 with plasma and at the instant t.

The ratio of carbon monoxide which was created during the discharge is defined by the following formulas:

$$R_{CO}(\%) = \frac{[CO](t)}{[CO_2]_0}\times 100 \tag{7}$$

Where $[CO](t)$ is the concentration of carbon monoxide at the instant t and N_T is the total density of the gas mixture.

3.1 Effect of the Applied Voltage

In Fig. 2, we have calculated the carbon dioxide conversion and carbon monoxide ratio under different values of applied voltage, for CO_2/O_2 (4%) gas mixture, frequency 50 kHz, gas pressure 500Torr, dielectric capacitance 230 pF, and gas temperature 300K.The obtained results indicate that the variations of these rates are slightly increased with the rising in the applied voltage. For voltage amplitude of 8 kV, the CO_2 conversion reaches a typical value of about 0.036%.

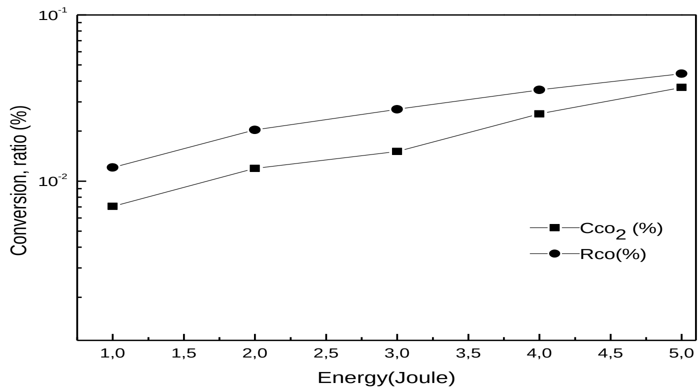

Fig. 2. Variations of carbon monoxide ratio and carbon dioxide conversion versus the energy deposed in the plasma.

3.2 Effect of the Frequency

In order to see the influence of discharge frequency on the time evolutions of discharge behavior, we performed calculations in this subsection for the following pa rameters: V_{app}= 6 kV, C_d = 230 pF, gas pressure 500 Torr and gas temperature 300 K, and 4% of O_2 in CO_2.We have varied the frequency from 20 to 200 kHz. This range of frequency is appropriate in this study. The results reported in Fig. 3 represent the variation of CO_2conversion and the CO ratio versus the discharge frequency. In addition, the ratio of CO is almost remains constant during the variation of frequency value. Moreover, the conversion of CO_2 is directly proportional to the frequency and its growth is quick at the discharge beginning (until 100 kHz). This conversion takes the greater value at highest frequency (200 kHz).

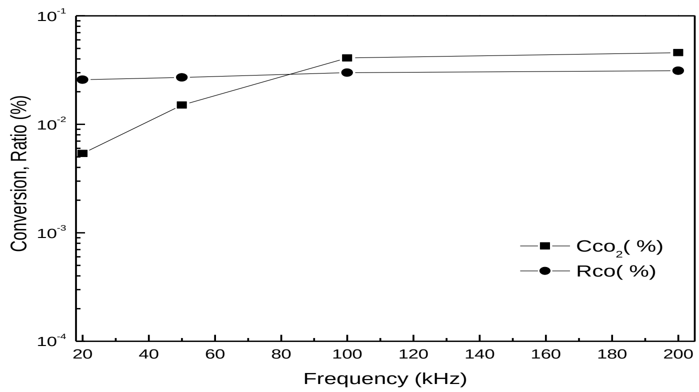

Fig. 3. Variations of carbon monoxide ratio and carbon dioxide conversion versus the frequency.

3.3 Effect of the Dielectric Capacitance

The calculations presented in this subsection are carried for the room temperature, frequency 50 kHz, gas pressure 500Torr, Vapp = 4 kV, and 4% of O_2 in gas mixture.

In Fig. 4, we plotted the conversion of CO_2, CO ratio versus the dielectric capacitance. The evolutions of these rates are directly proportional to the increasing in the dielectric capacitance value. The CO_2 conversion takes the most value of around 0.145% at 2000 pF

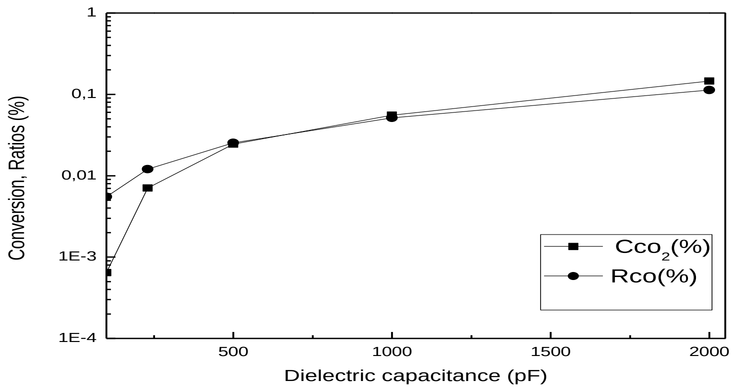

Fig. 4. Variations of carbon monoxide ratio and carbon dioxide conversion as a function of the dielectric capacitance.

3.4 Effect of the Gas Pressure

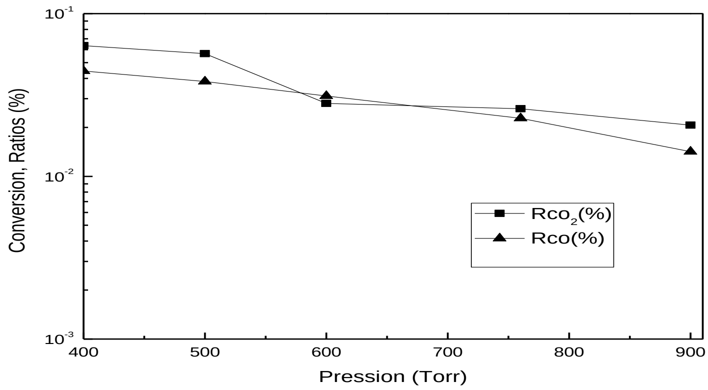

Fig. 5. Variations of the carbon monoxide ratio and carbon dioxide conversion as a function of the total gas pressure.

The effect of the gas pressure under the following conditions: frequency 100 kHz, Vapp = 7 kV, Cd = 230 pF, gas temperature 300 K, and 4% of O_2 in CO_2, on CO_2 conversion and CO ratio is presented in Fig. 5. We clearly see that the conversion of CO_2 and CO ratio decrease with the increasing of the gas pressure value. The most conversion of CO_2 was gained at lower pressure (400 Torr). For this pressure, the conversion of CO_2 is about 0.063%.

4 Conclusion

This work presents an electric and kinetic approach to study a homogeneous pulsed discharge in CO_2/O_2 gas mixture with typical operating conditions. It is based on a spatially homogeneous model (zero-dimensional model). Also, it is shown the effects of different plasma processing parameters such as applied voltage, gas pressure, dielectric capacitance, concentration of molecular oxygen in carbon dioxide, and frequency, on the discharge behavior. As well we have analyzed the effect of these parameters on the CO_2 conversion and the CO ratio. According to the calculated results and presented in this paper, on may conclude the following:

1. Higher conversion of CO_2, and CO ration can be obtained by increasing in the values of applied voltage, dielectric capacitance, and/or decreasing gas pressure.
2. CO_2 conversion is directly proportional to the rise of discharge frequency.

3. The discharge frequency indicates a weak influence on the ratio of carbon monoxide. In addition, CO_2 conversion is strongly affected by the dielectric capacitance.

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