Streamer modeling in air-based mixture

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Abstract

Non-thermal atmospheric pressure plasma technologies are more and more present in industrial processes. In particular, in pollution control processes where they have been shown efficient to treat a great variety of pollutant molecules in gas fluxes. To increase these processes efficiency, it is necessary to understand fundamental discharge mechanisms. Among all the discharges used, some of them present a filamentary aspect: dielectric barrier discharge, corona discharge. We present in this paper results concerning the modeling of the discharge process which occurs in a dielectric barrier discharge in air-based mixture. The three different phases of the discharge are presented (primary avalanche, streamer and post discharge). We particularly interest to the electric field evolution and to the primary chemical radical production.

1 Introduction

During the last century, pollutant gas exhausts in the atmosphere have increased considerably. The consequences are numerous in particular for human health and for the environment itself (ozone hole, global warming ...). To avoid these consequences, it is necessary to limit the toxic gas molecule exhaust. Important efforts have been made to decrease the pollutant molecule production, for example automobile industry has developed engine working at lower temperature to limit nitrogen oxides (NOx). But, in major cases it is not possible to avoid the pollutant molecules production, thus the gas must be treated before it is released in the atmosphere.

Treatment processes have been developed during the past years. Among all of them, a new technologies based on non-thermal plasma has appeared in the middle of the 80's. These technologies have been shown to treat variety of pollutant molecules in air-based fluxes, even for very dilute concentration.

The plasma produced in these discharges is non thermal in the sense of the majority of input energy is communicated preferentially to the electrons rather than heavy particles. Thus, these energetic electrons will be responsible of pollutant molecule destruction. The removal mechanism of pollutant molecules can take two ways. First, electron collides directly with the undesired molecules. This way is important for very concentrated pollutant molecules or for molecules with a high electron negativity affinity [2]. For example, very few ppm of CC4 in air react with electrons due to electron negative character of these electro negative atom. The second way occurs when the pollutant molecules are very diluted and have no electron negative affinity. In this case electrons collide principally with the background gas molecules (i.e. N2 and O2 for air) where they produce free chemical radicals. Then, these chemical radicals will induce a "classical" chemical kinetic.

In this paper, we particularly interest to the fundamental processes which occur in a dielectric barrier discharge reactor. We work with an air mixture. We have calculated the evolution of the different species produced during the discharge. We have also computed the electrical field distribution.

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2 Description of the model

Filamentary discharges are three dimensional processes (3D). Solving 3D equation set is too complex and very time consuming. That is why we have chosen to use a cylindrical model for the discharge. Furthermore we assume that the density of all species is uniformly distributed on the radius of the discharge. We use a fluid model to solve the transport equations. The space distribution of the charge in the gap disturb applied electrical field. Even if we work with a 1D model, we have to use, at least, a 2D description to compute electrical field [5]. The use of a 1D model for the transport equations and a 2D one for the electrical field is resumed as a "1.5D" model.

Our model describes the discharge that occurs in a dbd presenting a plane configuration. Only one electrode is covered by a dielectric (see figure 1). Cathode corresponds to the metallic pin and anode to the electrode covered with glassess. The gap g and the dielectric thickness are respectively of 3.75 mm and 1.5 mm.

The discharge cylinders has a radius of 200 μm. This value has been determinate by the work of Dhali [6]. Furthermore we suppose that when the discharge arises the inner face of the dielectric, electrons accumulate on a 500 μm disc. In this model, we assume that the discharge process is isothermal, during all the steps of the temperature remains equal to the room temperature (T = 300 K). This assumption has been verified with dbd made with a low electrical permeability dielectric [4]. Indeed, the temperature increase seems to be significant for dielectric with a permeability value above 50. And, we also suppose that the pressure is constant and equal to the 1 atm.

3 Fundamental equations

3.1 Equations

In this part, we present the fluid equations that describe the discharge. At room pressure, the mean times necessary to the equilibrium of densities, moments and energy are very small compared to the macroscopic parameters. A single moment description is so usual. For example, Braun [8] has computed the relaxation time corresponding to the diffusion and the dissociation of oxygen by the following mechanism : \( O_3 + e \rightarrow O^2P + O (1D) \). The authors have found that the electrons reach the equilibrium with the electrical field in about 5 ps. With this value and assuming a drift velocity of \( 10^5 \text{ m/s} \) for the electrons, we obtain a characteristic length of \( 5 \mu \text{m} \). This length is very small compared to the discharge dimension.

However, in works achieved by Wu and al [9], the authors have compared the results between a streamer computed with a fluid model in equilibrium and non equilibrium. They have shown that the ionization coefficient is in non equilibrium in the streamer head. Then, the streamer in the non equilibrium state is moving faster than the equilibrium one (15%).

The fundamental equations which describe the discharge behavior are the continuity equations (for electrons (1) and inline (2,3)) and the Poisson equation (4).
\[ \frac{\partial n_e}{\partial t} + \nabla \cdot (n_e \mathbf{v}_e) - \nabla \cdot \left( D_e \nabla n_e \right) = (\alpha - \eta) n_e |u_e| + s \]  
(1)

\[ \frac{\partial n_+}{\partial t} + \nabla \cdot (n_+ \mathbf{v}_+ - \nabla \cdot \left( D_+ \nabla n_+ \right) = \alpha n_e |u_+| + s \]  
(2)

\[ \frac{\partial n_-}{\partial t} + \nabla \cdot (n_- \mathbf{v}_-) - \nabla \cdot \left( D_- \nabla n_- \right) = \eta n_e |u_-| \]  
(3)

\[ \mathbf{v}_e \mathbf{E} = \frac{e(n_e + n_- - n_+)}{\varepsilon_0} \]  
(4)

where \( n_e, D_e, u_e, n_+, D_+, u_+, n_-, D_-, u_- \) are, respectively, the density of particles, diffusion coefficient and the drift velocity in the electrical field for the electrons, the positive ions and the negative ions. \( \alpha \) and \( \eta \) are the ionisation and attachment coefficient for the electrons. The reaction rates indicate the production or destruction by length unit for a particular specie.

The \( s \) factor which appears in equations (1) and (2) is a source term which represents the production of electrons (and positive ions) in the discharge. This electron production is necessary to assure the streamer propagation, in particular in its head. One of the possible secondary electron production mechanisms in the discharge is the photoionisation. Unfortunately, the knowledge of this phenomenon is only partial [1]. To avoid this problem, many authors [1,7] have taken into account photoionisation in their model in introducing a uniformly ionised background, fixed at \( 10^7 \) electrons per cm\(^3\). Many studies have been published to discuss the influence of this background on the streamer propagation.

Every coefficient \( (D, \alpha, \eta, \nu, s) \) which appears in the equation (1) to (2) are defined as tabulated as a function of the reduced field \( e = E/n \), where \( n \) is the total particle density. Their value can be determine by solving the Boltzmann equation with the collision cross sections [3]. The coefficients which appear in the equation are only a function of the reduced field because of local equilibrium.

3.2 Boundary conditions

To solve the equation system we used the boundary conditions used by Braun [3]. These conditions are:

- Secondary electrons are emitted from the cathode by ion collision on the cathode and by photo emission according to the formula
  \[ n_e (0, t) = \gamma_{\text{photo}} \int_{0}^{d} \alpha (x, t) u_e (x, t) n_e (x, t) \Omega (x) \, dx + \frac{\gamma_{\text{ion}}}{u_e (0, t)} n_+ (0, t) u_+ (0, t) \]  
(3)

Where \( \Omega \) represents the solid angle where the cathode is seen from the position \( x \). The parameter \( \gamma_{\text{photo}} \) at \( \gamma_{\text{ion}} \) represents the second Townsend coefficient, corresponding to ions and photons, respectively. Unfortunately, their value are not well determine experimentally. They are chosen equals and they are fixed to guaranty that the Townsend breakdown criteria is fulfilled [3].

- Ionic species are totally absorbed when they reach the cathode or the dielectric.

- When the electrons reach the dielectric: they accumulate homogeneously on the \( R_e \) radius circular surface.

4 Results and discussion

We have solved the equations above. The results are presented on the figure 2.

As we can see on the figure 2-a for the time between 0 and 60 ns, during the avalanche phase, electronic density (and ions) grow with the progression to the dielectric. After 60 ns, the space
Figure 2: Evolution of the discharge parameters -
a) Electrical field  
b) Electronic density  
c) Atomic nitrogen density  
d) Atomic oxygen density
charge density reach a sufficient value to start to disturb the applied field (Figure 2-a). At 85 ns, the distortion is becoming important: the streamer appears. It begins its progression towards the cathode (time from 94 ns to 99 ns). During its progression towards the cathode, its velocity and its intensity increase. The streamer leaves behind it a very weak electrical field (~ 100 Td). At t = 100 ns, the maximum of electrical field reach the cathode. The electrical field in the head of the streamer is, at this moment, about 900 Td. This corresponds to electrons with an energetic value close to 14 eV.

Then begins the post-discharge phase which corresponds to the regime of glow discharges. We can see a very strong cathodic drop, and after a space zone where an uniform electrical field exists. Its strength decreases with time from 100 Td to 20 Td. This electrical field valueinduces electrons with a mean energy of about 1 eV. As we can see on figure 2-a, electrical field strength decrease while the electrons accumulate on the dielectric. A t = 120 ns, the field has a value of 20 Td: the discharge is extinguishing.

The Figure 2-c and 2-d represent respectively the density of atomic nitrogen N and atomic oxygen O. As we can see, it is during the streamer phase that the radical production is the most important. When the streamer has reached the cathode, the density values are $2 \times 10^{18}$ and $1.5 \times 10^{18}$ cm$^{-3}$ for the nitrogen and oxygen atoms. At the end of the avalanche phase, these densities are only $3 \times 10^{18}$ and $5 \times 10^{14}$ cm$^{-3}$. Due to the high strength of the electrical field in the streamer, the chemical radical (N and O) production is increased twice.

5 conclusion

In this paper, we have computed a 1D streamer model. Even if the assumptions we made were simplifying, this method allows us to determine the density of free chemical radicals produce at the end of the discharge. This work is the first step for a complete chemical model used to compute the chemical kinetics in plasma assisted pollution control processes.

References
