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Atmospheric pressure plasma jet based on the dielectric barrier discharge

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Abstract. The dielectric barrier discharge has attracted the interest of many researchers in several fields since it produces a non-thermal plasma at atmospheric pressure. Its field of application is totally vast and includes medicine, biotechnology, chemistry, and various other fields of science and technology because of the physical and chemical properties that characterize the plasma jet based on the dielectric barrier discharge. These are characterized by different possible configurations and can be powered by radio frequency, alternating current, and even pulsed direct current depending on the application. In this work, we have presented the atmospheric pressure plasma jet as a new micro discharge that can be used in several fields. We have presented the different possible configurations of the plasma jets based on the dielectric barrier discharge that vary according to the targeted application field. In addition, we have presented some fields of study and specialties in the different application areas such as medicine, biotechnology and food preservation, agriculture and combustion domains. Finally, we have presented the results of the simulation with COMSOL Multiphysics of a dielectric barrier discharge at atmospheric pressure for different frequencies and different discharge gaps between the electrodes, since this is the discharge used in the plasma jet.

1 Introduction

Atmospheric pressure plasma jets allow the non-thermal plasmas to be effectively formed and are easy to use in ambient air conditions since they are considered as new micro discharges. Numerous theoretical and experimental studies based on the chemical and physical properties of plasma jets at atmospheric pressure are carried out in various fields such as medicine, biotechnology, chemistry, nanotechnology, surface modification, and many other fields of science and technology [1][2][3][4]. Various types of gases can be chosen to generate cold atmospheric plasma like helium, nitrogen, argon, air, and heliox a mix of helium and oxygen [5]. Most of the researches reported in several journals has used a plasma jet propagating in a helium stream surrounded by free air at ambient temperature for its potential capacity to deliver high flows of reactive species to a target located a few centimeters away which is interesting in the different fields of application. In this paper, two simulations of a dielectric barrier discharge have been realized with a 2D model at atmospheric pressure using helium with an applied sinusoidal voltage of 1kV, in order to study the influence of the applied voltage frequency which varies between 200 Hz and 10kHz, and the influence of the discharge diameter between the electrodes which varies between 0.8 mm and 2 mm, on the gas breakdown voltage and on the discharge current intensity.

2 DBD plasma jet

The dielectric barrier discharge is an electrical discharge created between two electrodes separated by a dielectric material. In general, the discharge is created by an alternating current applied to the terminals of the electrodes. If a very high potential is applied, a breakdown voltage can be reached. When this voltage is attained between two simple electrodes, an electric arc is created between them. This arc is due to a concentration of charges localized in one place on the surface of an electrode. To avoid this arc, dielectrics are placed on one or both electrodes or even between the electrodes, the role of this dielectric is to distribute the charges accumulated on their surface. Moreover, by accumulating charges on their surfaces, the dielectric helps to create an electric field opposite to the electric field applied to the terminals of the electrodes. This last property stops the discharge before the transition to the arc at the voltage predicted by Paschen's law. Instead of having a very energetic and localized discharge, we will have a stable and distributed discharge on the total surface of the electrodes hidden by the dielectrics.[6][7].

Several types with different configurations of atmospheric pressure plasma jet have been referred to in different research works, most of them operating with a pure noble gas or mixed with a little portion of reactive gases, such as air. Plasma jets operating with noble gases can be classified into four categories [8][9], i. e.

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2.1 Dielectric-free electrode jets (DFEJ)

Dielectric-free electrode jet (DFEJ) is powered by a 13.56 MHz radiofrequency energy source. It is composed by an internal electrode, which is coupled to the energy source, and an external grounded electrode as depicted in Figure 1.

A mixture of He and reactive gases is introduced into the annular space between the two electrodes at 51L/min. Cooling water is required to prevent jet superheating, and the plasma jet gas temperature ranges from 50 to 300 °C, depending on the radio frequency power. The researchers found for this type of dielectric free electrode jet that the power delivered to the plasma is much higher which leads to a strong increase of the gas temperature and subsequently, the plasma will be very reactive which is not acceptable for biomedical applications. The electric field inside the discharge space and along the direction of propagation of the plasma plume is relatively weak since this jet is powered by a radio frequency. Thus, the propagation of this plasma plume is probably driven by the gas flow than by electricity. [5][10][11]

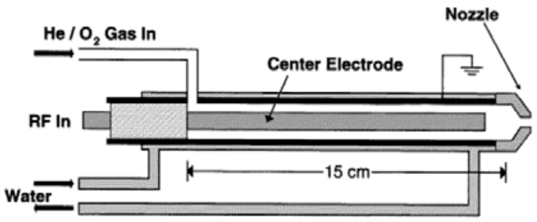


fig 1: Schematic of the dielectric-free electrode atmospheric-pressure plasma jet[11]

2.2 Dielectric barrier discharge jets (DBDJ)

Dielectric barrier discharge jets (DBDJ), characterized by several jet configurations with one or two dielectric barriers. Figure 2(a,b) represents the first configuration of dielectric barrier plasma jets which is characterized by a dielectric tube surrounded on the outer side by two metallic ring electrodes. When a high voltage of kHz is applied across the electrodes and a working gas (He or Ar) passes through the dielectric tube, a cold plasma jet is created in the surrounding air. This type was first described[12] and reviewed [5],[13]. The authors found that the plasma jet consumes a low power of a few watts. The temperature of the plasma gas is equivalent to the ambient temperature, and that the plasma jet, which appears to the naked eye to be homogeneous, is, in reality, a bullet-like volume of plasma with a high propagation Velocity higher than 10 km s^{-1}. By elimination of a ring electrode as shown in figure 2(e) the discharge inside the dielectric tube becomes weaker [14]. To improve the electric field along the plasma jet which is very favorable for generating longer plasma jet and for more active plasma chemistry, [15][16] replaced a high voltage ring electrode with a pin electrode centered inside a dielectric tube, as shown in Figure 2(c,d). If the second ring electrode is eliminated as shown in figure 2(f), the discharge inside the dielectric tube becomes weaker[15].

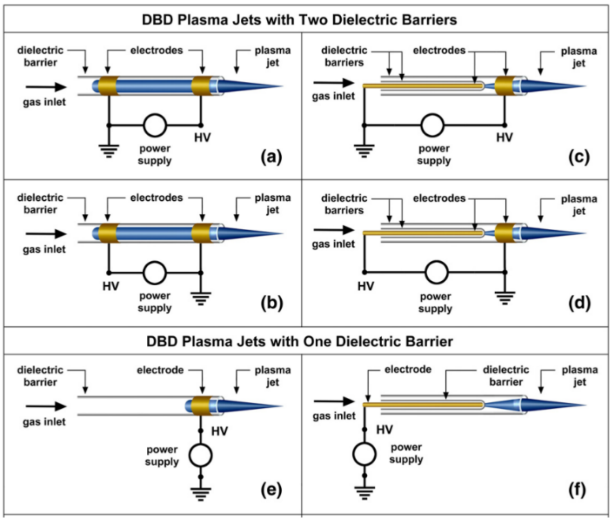


fig 2 : Different configurations of DBD plasma jet [13]

2.3 DBD-like plasma jets

DBD-like plasma jets in figure 3, are called DBD-like because, when the plasma jet is free of contact with a conductive object, the discharge looks approximately like a dielectric barrier discharge. Whereas if the plasma jet is in contact with a conductive object, the discharge passes between the conductive object and the high-voltage electrode, in this stage, it is not a dielectric barrier discharge anymore. Although other DBD systems do not present any risk of having an arc between the object and the dielectric whatever the distance between them, on the other hand for plasma jets of DBD-like there is a risk of having an electric arc depending on the material used is conductive or not and even depending on the field of application. If this arc is avoided, the use of this type presents many advantages [5] [17]. In addition, DBD plasma jets can be powered by radio frequency power, kHz AC power, and even pulsed DC power. The second configuration is to replace the high-voltage electrode with a hollow electrode as shown in figure 3(b), they found that the plasma jet is longer by injecting a noble gas in the first inlet and a reactive gas in the second inlet [18][19].

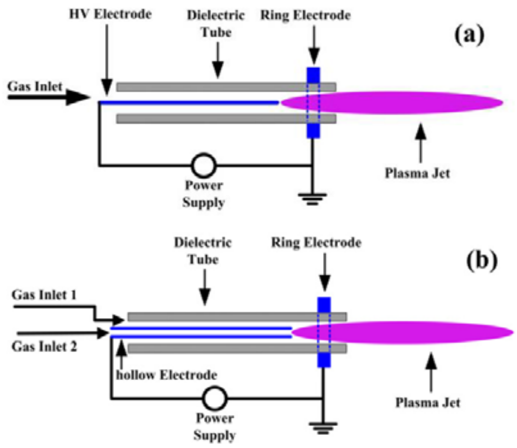


fig 3: Schematic of a DBD-like plasma jet.[5]

2.4 Single electrode jets (SE)

Single electrode jets (SE), Single electrode jets, or plasma needle jets, are basically the same as DBD-like jets, the only difference being that the ring electrode outside the electric tube is eliminated in this model, so the dielectric tube plays a unique role in directing the gas flow [20][21]. This type of jet can also be powered by direct current, kHz alternating current, radio frequency or pulsed direct current [22].

3 Research areas and applications

The applications of atmospheric pressure plasma based on the dielectric barrier discharge in several fields, depending on the technology used, have attracted the interest of many researchers. Among these fields of application, we find the decomposition of volatile organic compounds (VOCs) and the most well-known combustion gases such as butane, toluene, ethanol (90° alcohol), acetone and benzene, which are found in the industrial field during various industrial processes[23][24]. The conversion of greenhouse gases into liquid fuel [25]. The sterilization of water [26]. The treatment of gases and wastewater by the destruction of dangerous organic pollutants [24], the modification of surfaces and the treatment of soils and wastes. Plasma can also be used in medicine in order to treat several diseases, disinfection and sterilization [27]. In the agricultural field and food industry, plasma is used for germination of seeds, growth of plants, storage of products, sterilization of packaging, disinfection, pasteurization and food preservation[28][29].

3.1 Plasma medicine

Plasma medicine is an exciting field where life science and physics intersect. The ability to produce a cold plasma at atmospheric pressure, which is characterized by a multitude of charged particles, reactive gaseous species, electric current, and ultraviolet rays, has been the basis for the fast growth of plasma-related application areas in biomedicine [30]. Plasma can be used in oncology or cancerology; it has been demonstrated that plasma causes apoptosis or programmed physiological cell death of tumor cells as well as reducing skin tumors [31]. To this end, several dermatology studies have been conducted to investigate the efficacy and safety of cold plasma in dermatooncology. To date, plasma has been used for the treatment of eczema and has not caused any negative side effects, according to experiments conducted on more than 40 patients who were treated daily with plasma for two minutes [32][33]. Atmospheric pressure plasma is also used for disinfection, numerous studies have shown the lethality of plasma on fungi and bacteria[34], such as group A streptococcus, Escherichia coli, Pseudomonas Aeruginosa, and (MRSA) methicillin-resistant staphylococcus aureus, as well as the elimination of fungal strains clinically relevant by atmospheric pressure plasma[35][36][37] according to experiments realized on 19 patients with onychomycosis of the nails. Another use of atmospheric pressure plasma on human skin concerns the hydrolipidic film composed by several bacteria, produced by the sebaceous and perspiratory glands. This hydrolipidic film protects the skin against dryness with a pH value between 5.4 and 5.9 for healthy skin [38]. An increase in the pH of the hydrolipidic film leads to a change in the microbial load which causes diseases, such as atopic dermatitis and ichthyosis[39]. Since pH values above 6 are favored by pathogenic bacteria[40].

The interaction of the hydrolipidic film of diseased skin with chemical compounds in the plasma causes a decrease in pH values after five seconds according to experiments on patients with ichthyosis [41], the decrease in pH leads to inhibition of pathogens, and consequently, healing of the skin. Atmospheric pressure plasma helps to accelerate the healing of wounds[42][43] the treatment of scars and acne by decreasing nearly 80% of the sebum production [44]. Generally, the field of plasma medicine is very interesting and exciting, and numerous experiments provide the use of cold plasma for the treatment of various dermatological and oncological diseases.

3.2 Food preservation

The preservation treatments applied to food aim to preserve their edibility and their gustatory and nutritional properties by preventing the development of bacteria, fungi and microorganisms that they contain and that can in some cases lead to food poisoning. The methods used for the preservation of food are based on: heat (pasteurization, sterilization, appertization ...), cold (deep freezing, freezing, and refrigeration ...), there are also vacuum packaging or modified atmosphere, dehydration and drying, fermentation, salting, pickling, smoking, ionization, etc... Spores and spore-forming organisms, when exposed over a long period of time to these mentioned treatments, become resistant. Which prompted researchers [45][46], to develop new preservation technologies among them we find the use of plasma at atmospheric pressure based on dielectric barrier discharge. These studies were based on the influence of different parameters such as the type of electrode and the method of direct or indirect exposure as well as the distance from the sample, the power used, the dose and duration of the treatment and also the interaction of the reactive species of the plasma with bacteria and spores[47][48].

It has been proven that plasma preservation at atmospheric pressure has several advantages such as the simplicity of using the equipment in an ambient temperature and the rapidity of treatment, it improves the stability of food with better nutritional value and the reduction of microbial load [45][47][49].

3.3 Agricultural sector

Atmospheric pressure plasma can be used in the agricultural field. Several devices have been developed according to their application either at the pre-planting, pre-harvest or post-harvest level [50][51]. Atmospheric pressure plasma technologies can be used to disinfect crops or food after harvest [51], decontaminate seeds during storage, improve growth and germination of seeds, reduce soil invasion by pathogens by adding reactive species which leads to a decrease in pH, treatment and sterilization of water used for washing products[52], volatile organic compounds in storage installations and food preparation equipment, and finally the destruction of dangerous food residues.

3.4 Combustion sector

Dielectric barrier discharge is one of the most useful alternative methods for atmospheric pressure plasma combustion. With the aim of satisfying the urgent needs of having a clean and stable combustion, while preserving ozone, this combustion method is attracting attention in the last decades from several researchers[53][54]. Several parameters allow the dielectric barrier discharge to manipulate and control combustions, first, its simple structure and flexibility at variable pressures, second, by using different voltage waveforms and with adjustable frequencies and amplitudes the DBD causes significant ionic winds[55], furthermore, the DBD allows the modification of the kinetic pathway through the generation of radicals and active species [56][57]. Even at the configuration of the DBD used, [58], showed that the configuration plays an important role in the control and stability of the flame.

4 Modelization of plasma at atmospheric pressure

In order to understand the atmospheric pressure plasma based on the dielectric barrier discharge, a two-dimensional model has been used at atmospheric pressure (760 Torr) using Helium as a noble gas, with sinusoidal voltage, a discharge gap between two electrodes varying between 0.1 mm and 2 mm, a relative dielectric permeability of 10. This dielectric permit to avoid the concentration of localized charges in a single place on the surface of the electrodes and subsequently it allows to avoid the creation of an arc between the electrodes.

fig 4: simulation model

In order to calculate the different physical characteristics such as electron density, energy density of electrons. the local energy approximation is used which is based on the solution of a system that contains three fluid equations called moment of Boltzman equation.

$$\frac{\partial(n_e)}{\partial t} + \nabla\,\Gamma_e = R_e \qquad (1)$$

$$\Gamma_e = -n_e(\mu_e.\boldsymbol{E}) - \boldsymbol{D}_e.\nabla n_e \qquad (2)$$

With n_e: electronic density (1/m3). μ_e: electron mobility (m2/ (V.s)). R_e: electron rate (1/ (m3. S)). $\boldsymbol{D}_e$: electron diffusivity. E: electric field (V/m). $[-(\mu_e.\boldsymbol{E})n_e]$: indicates the electron migration due to an electric field. $(-\boldsymbol{D}_e.\nabla n_e)$: the diffusion of electrons from high to low electron density regions.

$$\frac{\partial(n_\varepsilon)}{\partial t} + \nabla\,\Gamma_\varepsilon + \boldsymbol{E}.\Gamma_e = R_\varepsilon \qquad (3)$$

$$\Gamma_\varepsilon = -n_\varepsilon(\mu_\varepsilon.\boldsymbol{E}) - \boldsymbol{D}_\varepsilon.\nabla n_\varepsilon \qquad (4)$$

With n_ε: electron energy density (V/m^3). μ_ε: energetic mobility of electrons (m^2/(V.s)). R_ε: Loss/gain of energy due to inelastic collisions. $\boldsymbol{D}_\varepsilon$**:** energetic diffusivity of electrons. $\boldsymbol{E}.\Gamma_e$: the electrons heating due to external E, it can be a heat source or a heat sink, depending on if the electrons drift or not in the same direction as the electric field.

The energy diffusivity, the energy mobility and the electron diffusivity are calculated from the electron mobility using the Maxwellian electron energy distribution function.

$$\boldsymbol{D}_e = \mu_e T_e \qquad (5)$$

$$\mu_\varepsilon = \left(\frac{5}{3}\right)\mu_e \qquad (6)$$

$$\boldsymbol{D}_\varepsilon = \mu_\varepsilon T_e \qquad (7)$$

T_e: temperature: is a function of the mean electron energy, ε

The accumulation of charges on the dielectric surface leads to the following boundary condition:

$$n.(\boldsymbol{D}_1 - \boldsymbol{D}_2) = \rho \qquad (8)$$

$$n.(\boldsymbol{E}_1\,\boldsymbol{\varepsilon}_1 - \boldsymbol{E}_2\,\boldsymbol{\varepsilon}_2) = \rho \qquad (9)$$

$\boldsymbol{\varepsilon}_1$ and $\boldsymbol{\varepsilon}_2$: the relative permittivities of the dielectric and the gas, respectively.

The chemical reactions taken into consideration are respectively of the types: Elastic, Excitation and Ionization.

e+He => e+He (10)

e+He => e+Hes (11)

e+Hes => e+he$^+$ (12)

5 Results

The high-pressure discharge with a higher species density and a fairly important space charge generates more interactions between the particles and a low mean free path. hence the considerable electric field plays an important role in the breakdown of the gas. The mechanism involved in this case is called "Streamer mechanism".

The electrical characteristics calculated when two cycles of voltage are applied at atmospheric pressure (760 torr) with a discharge distance equal to 0.5 mm are shown in Figure 5 and 6 at different frequencies (a) 200Hz, (b) 1kHz and (c) 10kHz.

We notice from the simulation results obtained in fig 5 that the increase of the frequency of the applied voltage causes an increase of the intensity of the discharge current which varies from 15.10^{-6} (A) for a 200Hz frequency to 9.10^{-5} (A) for a 1kHz and then the current intensity reaches 0.003 (A) at 10kHz.

These results can be explained by the fact that when we increase the frequency the period between two maximums is reduced and then the applied voltage reaches the breakdown voltage quickly which reduces the discharge rate which is illustrated on the figures by the number of peaks which is decreased, and finally, we pass from the filamentary regime to a homogeneous regime.

From fig 6 we find that the variation of positive ion density varies in the same way as that of the discharge current in fig 5 with the same peaks, which is totally natural, explained by the fact that the addition of the dielectric stops the accumulation of charges on the surface of the electrodes to avoid the creation of an electric arc. Then it creates an electric field in the opposite direction to the applied electric field. So, the ions propagate in the direction of the applied electric field and the electrons propagate in the opposite direction which requires that the electrons and the ions must be created in an identical way to preserve the charge balance, which explains why the peaks are identical.

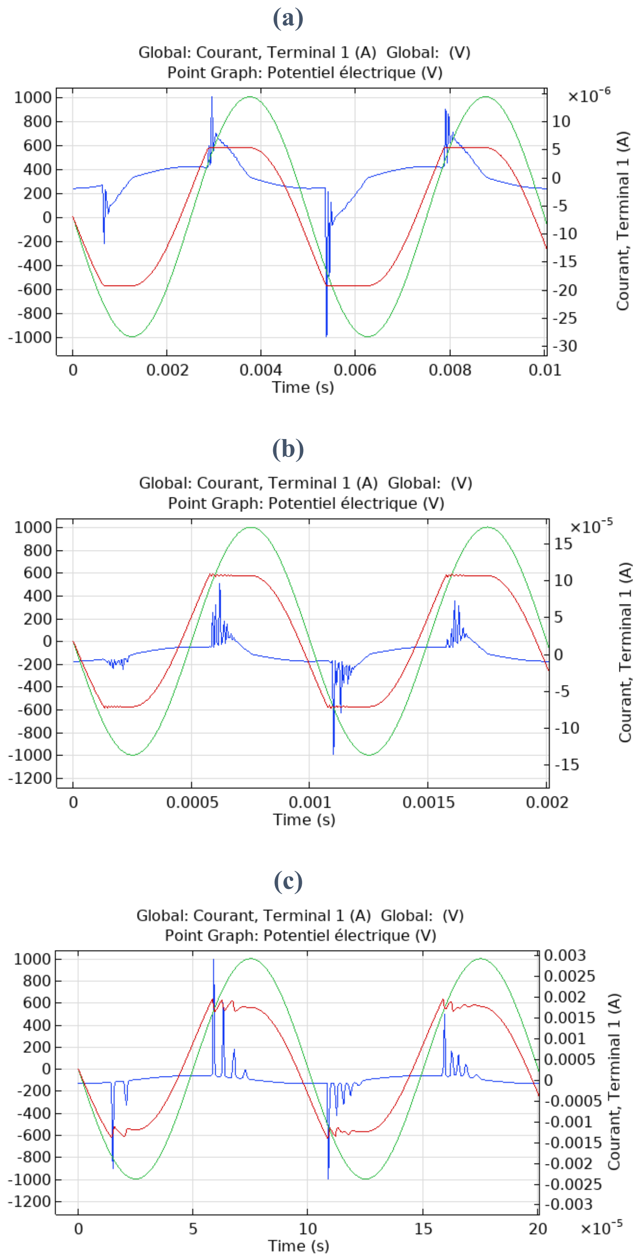


fig 5 : the variation of the breakdown voltage (in red) and of the electric current (in blue) as a function of the applied voltage (in green) with d = 0.5 mm and P = 760 Torr at different frequencies (a) 200Hz, (b) 1kHz and (c) 10kHz.

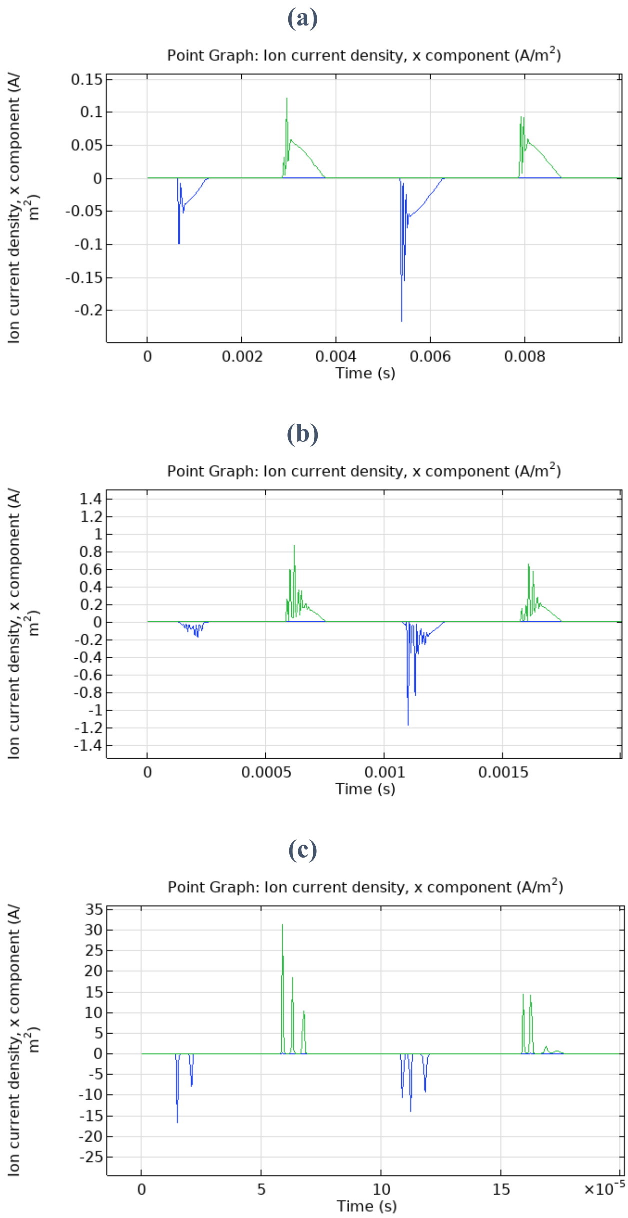


fig 6 : the variation of positive helium ion density, (in blue) next to the dielectric and (in green) next to the cathode with d = 0.5 mm and P = 760 Torr at different frequencies (a) 200Hz, (b) 1kHz and (c) 10kHz.

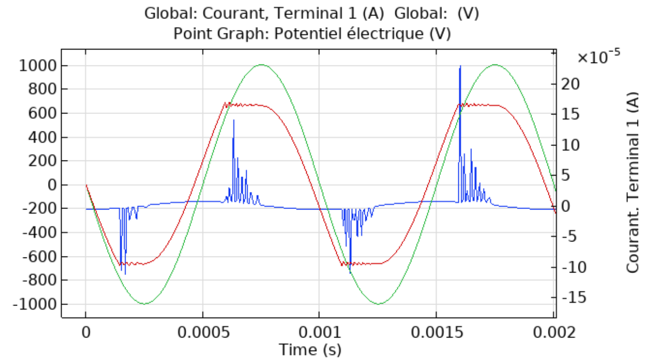
On fig 7 and 8 another study is presented to better understand the influence of the distance of discharge which separates the electrodes on the intensity amplitude of the discharge current, while keeping the same frequency on the applied voltage. The results of simulation on fig 7 show that with the enlargement of the distance of discharge, the intensity amplitude of current decreases from 25.10^{-5} for a distance of 0.8 mm to 6.10^{-5} for a distance of 2mm passing by 15.10^{-5} for 1mm of distance. This diminution of the discharge distance provokes a reduction of the number of peaks.

These results can be explained by the fact that with the same applied voltage and the same frequency when the discharge distance is very small the particles inside are very close which generates more interaction between them which causes an increase in the number of peaks and an increase in the intensity of the discharge current. and inversely, more the electrode moves away more the particles move away too, which makes the interactions between the particles weaker and consequently decreases the number of peaks.

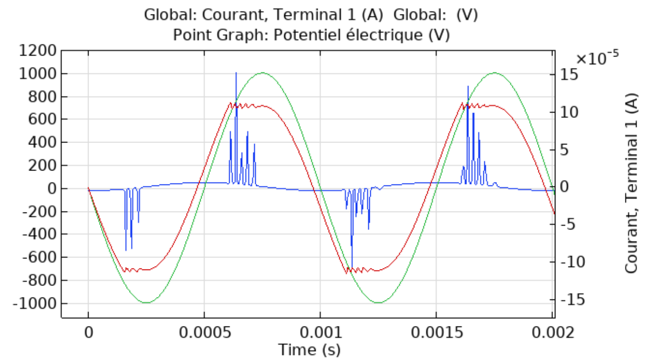
The simulation results on figure 8 concerning the variation of ions density compared by the results presented on figure 7 confirms what we concluded previously that ions propagate in the direction of the applied electric field and electrons propagate in the opposite direction which requires that electrons and ions must be created in an identical way to preserve the charge balance.

These obtained simulations results are in agreement with several experimental studies performed which presented the variation of discharge current intensity and applied voltage for different frequencies: 500 Hz [21], 18 kHz[31], 27 kHz [30], 10 kHz [59].

(a)



(b)



(c)

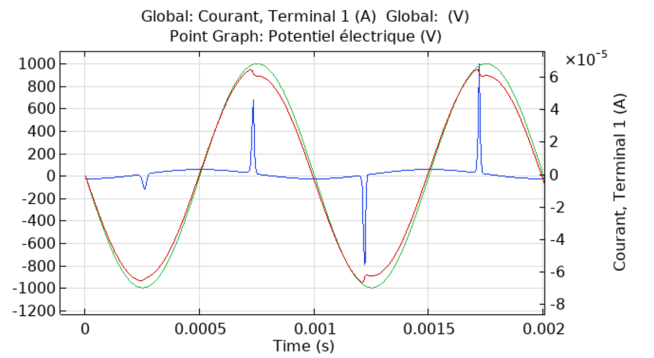
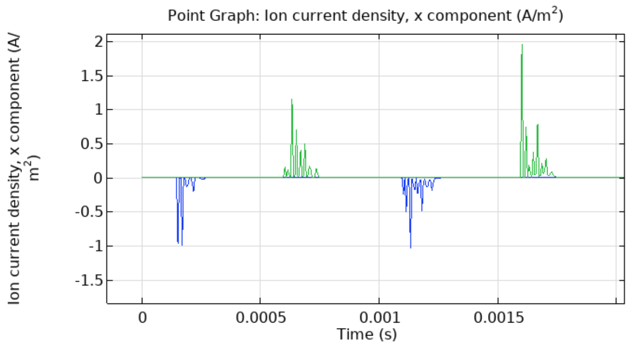


fig 7 : the variation of the breakdown voltage (in red) and of the electric current (in blue) as a function of the applied voltage (in green) with f = 1kHz and P = 760 Torr at different discharge distances (a)0.8mm, (b) 1mm and (c)2mm.

(a)



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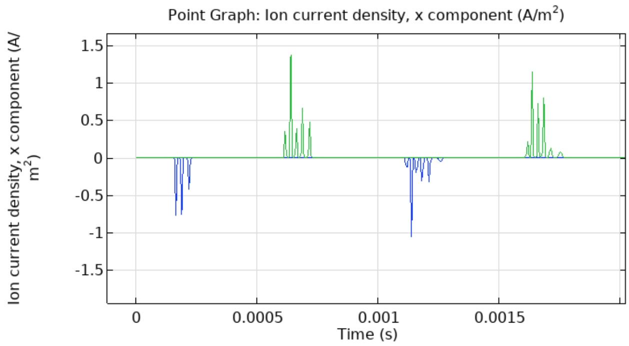

(c)

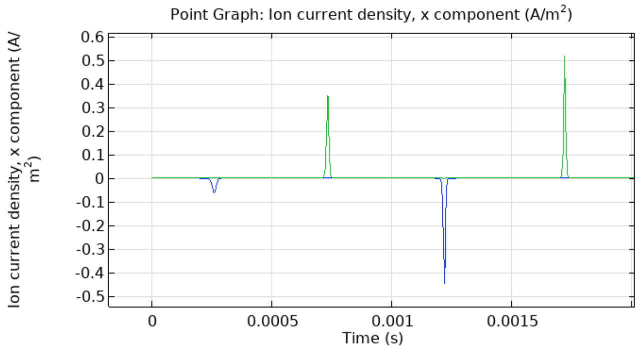

fig 8 : the variation of positive helium ion density, (in blue) next to the dielectric and (in green) next to the cathode with f = 1kHz and P = 760 Torr at different discharge distances (a)0.8mm, (b) 1mm and (c)2mm.

6 Conclusion

The atmospheric pressure plasma based on the dielectric barrier discharge can be used in different fields and with different configurations depending on the application desired due to its physical and chemical properties. In this work two numerical studies were carried out using a two-dimensional model with helium inside with a sinusoidal voltage applied of 1kV.

The first study was dedicated to determine the influence of the applied frequency on the gas breakdown voltage and on the intensity of the discharge current, keeping the same distance of discharge between the electrodes. It was concluded that the increase of the frequency provokes an increase of the current intensity amplitude, but the number of peaks decreases and consequently the rate of discharge decreases as well.

The second study was allocated to determine the influence of the discharge distance between the electrodes on the intensity amplitude of the discharge current while keeping the same frequency on the applied voltage. It was concluded that the widening of the discharge distance decreases the intensity of the discharge current and minimizes the number of peaks. Therefore, from these two studies we conclude that we can go from a filamentary or non-homogeneous regime to a homogeneous one, either by widening the discharge distance or by increasing the frequency of the applied voltage.

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