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Non Thermal Plasma Sources of Production of Active Species for Biomedical Uses: Analyses, Optimization and Prospect

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1. Introduction

Non thermal and low temperature plasmas in air or in another gas or gas mixtures at atmospheric pressure or at reduced pressure are very efficient sources of active species (radicals, excited species, charged particles, photons emission covering UV up to IR wavelength range, etc.). The case of reduced pressure plasmas where the gas is conditioned into a tight cell with no contact with ambient air can be very interesting for the treatments of more particularly inert material and also for in vitro plasma exposure when it is needed a well controlled gas composition and specific active species for instance without molecular oxygen in order to avoid oxidation processes. The atmospheric pressure plasma in comparison to the lower pressure cases, can be directly used in the ambient air without using any vacuum system and no gas feed but the nature of the active species are not well controlled because this includes many byproducts of the main molecules of air (N_2 and O_2) and also the byproducts of the low concentration species or impurities (H_2O , CO_2 , NO_x , ozone, rare gases, etc.). Many applications can be fulfilled from such non thermal plasmas in for instance the domains of gas pollution control (see e.g. Penetrante 1993, Eichwald et al 1998, Dorai 2003, Kim 2004) or surface treatment (see e.g. Kunhardt 2008, Fang 2004 and Bhoj 2008), or plasma actuators (see e.g. Eichwald et al 1998, Moreau 2007). There are also the biomedical applications of low temperature plasmas concerning sterilization and decontamination (see e.g. Laroussi 2002, Villeger et al 2005, Pointu et al 2005, Sarrette et al 2010), engineering of tissues and biomaterial (see e.g. Sardella et al 2008, d'Agostino 2008, Desmet et al 2009) and more recently the field of plasma medicine (see e.g. Fridman et al 2008, Stoffels et al 2008, Morfill et al 2009, Lloyd et al 2010, Wetman et al 2010, Kim et al 2010).

In the sections following this introduction, an overview on several kinds of non thermal and low temperature plasma sources are given in section 2 with a specific emphasis on sources of active species used in our laboratory. Sections 3 and 4 are devoted to the characterization and the possible optimizations of these plasma sources of active species by using experimental diagnostics as well as the different discharge modeling tools experimentally validated and fed by basic data collected or determined in the case of the non thermal

plasmas of our interest. In section 5, two illustrative examples of the use of active species at reduced and atmospheric pressure are shown in the field of biomedical applications.

2. Overview on different non thermal plasma sources

An overview on the main kinds of plasma sources that can be used more particularly for biomedical applications will be given in this section with a focus on the plasma sources used in our laboratory (<http://www.laplace.univ-tlse.fr/groupe-de-recherche/plasmas-reactifs-hors-equilibre/>)

2.1 Flowing afterglow discharge generated at reduced pressure by a microwave source for plasma remote sterilization

In the experimental setup developed in our laboratory for sterilization applications (see e.g. Villeger et al 2005, Ricard et al 2007, Sarrette et al 2010), the discharge is generated by a classical 2.45 GHz microwave surfatron cavity at powers up to 300 W. The microwave plasma is then launched using a gas flow through a small diameter tube up to a Pyrex treatment chamber of 15 cm diameter and 20 cm height. The gas flow could be varied up to 3 l/min and the gas pressure in the reactor is adjustable between 0.1 and 5 kPa by means of a throttle valve above the primary pump. Such conditioned gas at reduced pressure allows the use of different gas compositions as for instance pure N_2 , and also gas mixtures such as Ar/N_2 , H_2/N_2 , or O_2/N_2 following the species required for the sterilization processes as for instance the atomic nitrogen whose the recombination at the surface of the bacteria (see support in Fig. 1a) plays a major role during the sterilization process. In fact, the reduced pressure has the advantage to avoid the volume recombination of atomic nitrogen in the flowing afterglow and also to need a low surfatron power.

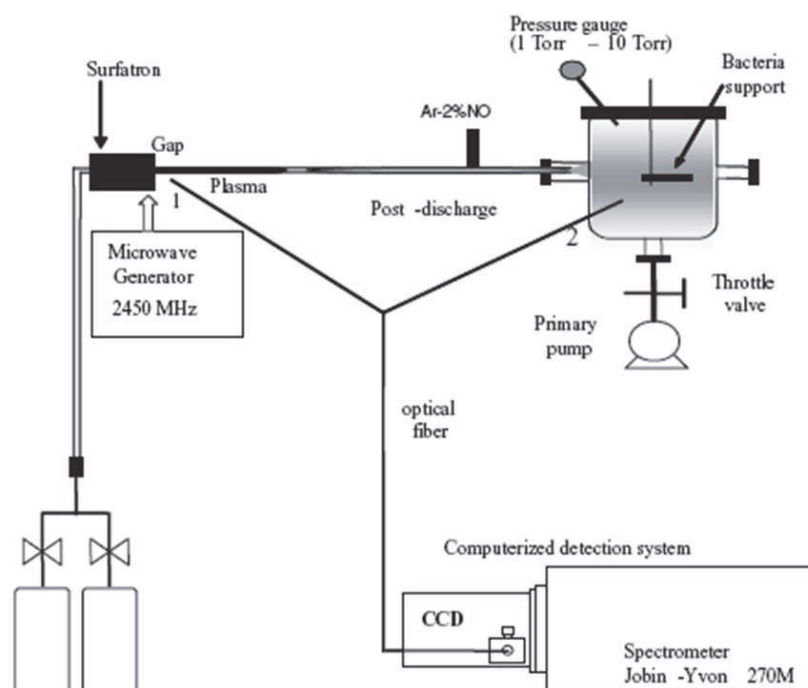


Fig. 1. a. Experimental set up generating a flowing afterglow discharge for sterilization (from Ricard et al 2007).

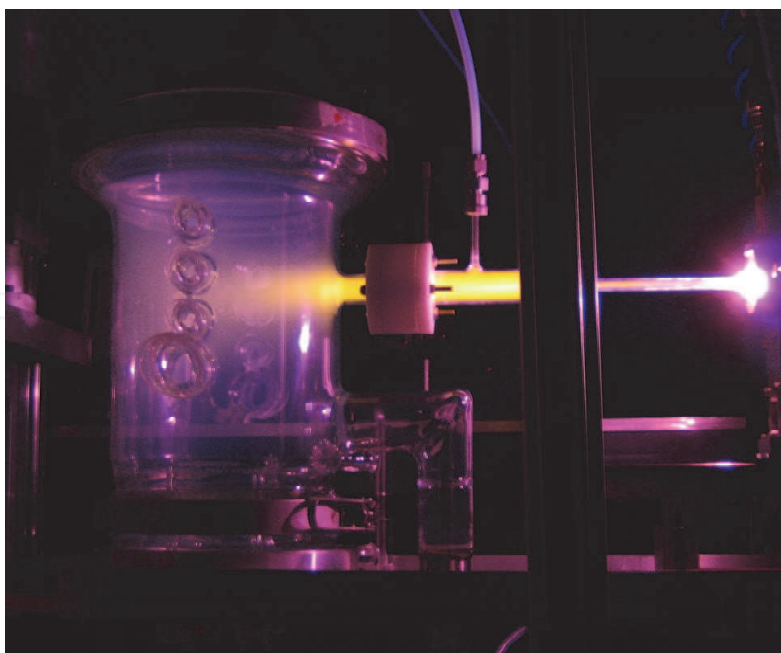


Fig. 1. b. View on the experimental setup generating flowing afterglow discharge (surfatron is on the right side and the treatment chamber on the left side)

2.2 DBD discharge using a mono-polar source

Dielectrics barriers discharges (DBD) are very well known since a long time with one or two electrodes covered by the dielectric (Fig. 2). The role of the dielectric is to limit the current thus avoiding the gas spark or breakdown. Under such configuration, an alternating or pulsed power supply is generally used in the case of a quite low (a few mm) gap distance (see e.g. Eliasson 1998, Koslov 2005). Generally the DBD power supply is a bipolar source. The polarity inversion at the half period allows to the discharge, stopped by the charge accumulated on the dielectric, to start again. It is worth noting that a mono polar pulsed power was successfully tested in the literature (Liu et al 2001, Laroussi et al 2004) and in our laboratory (Panousis et al 2009). In the case of such a mono-polar power supply, the discharge occurs alternatively during the increasing part of the pulsed voltage and is inverted during the decreasing one. It is shown that this mono polar system leads to better efficiency of the production of active species than a classical bipolar power supply.

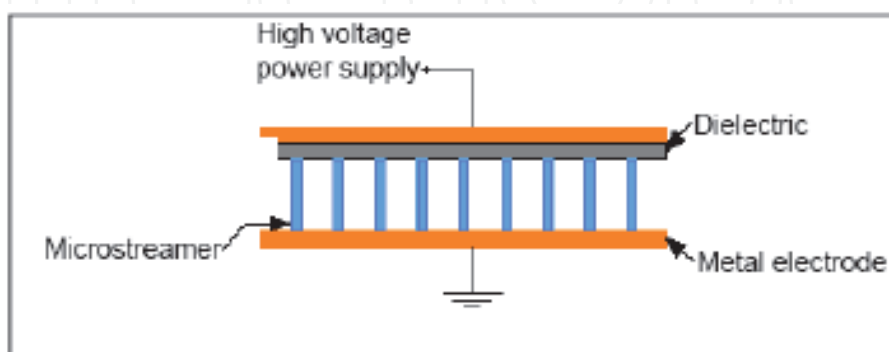


Fig. 2. Schematic view of a DBD discharge with a barrier dielectric on the upper electrode and a filamentary structure which is its usual morphology at atmospheric pressure

2.3 Volume corona discharges for mono and multi-tips

Corona discharges can be generated under many electrode configurations, provided one of which has a curved shape as a tip or a wire, etc. In the case of for instance a positive point-to-plane geometry under DC or pulsed power supply, the discharge can have either a mono-filamentary structure for DC power supply or a branching structure for pulsed power supply (see e.g. Merbahi 2008, Briels et al 2005). Fig. 3a displays the mono-filament and branching cases. The filamentary structure corresponds to a complex streamer dynamics that is analyzed largely in the literature showing that the active species are generated inside the ionized channels during both primary and secondary streamer developments (see e.g. Eichwald et al 2008). Furthermore in order to increase the efficiency of the production of active species it is better to use a multi-tip electrode configuration (Fig. 3b) where it is easy to see in Fig. 4, showing the instantaneous discharge current, that the peak of the corona current and therefore the dissipated power does not increase linearly with the number of the tips.

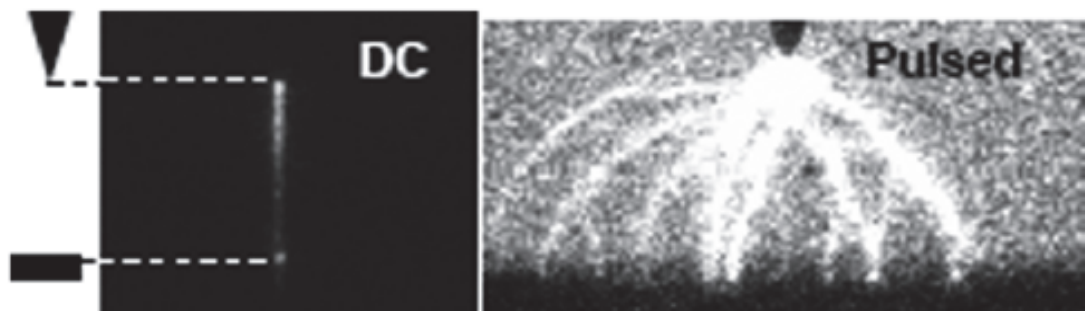


Fig. 3. a. Dc and pulsed corona discharge in a positive point-to-plane geometry (from Abahazem et al 2008)



Fig. 3. b. View on a multi-tip pulsed corona discharge tested at LAPLACE-PRHE Laboratory.

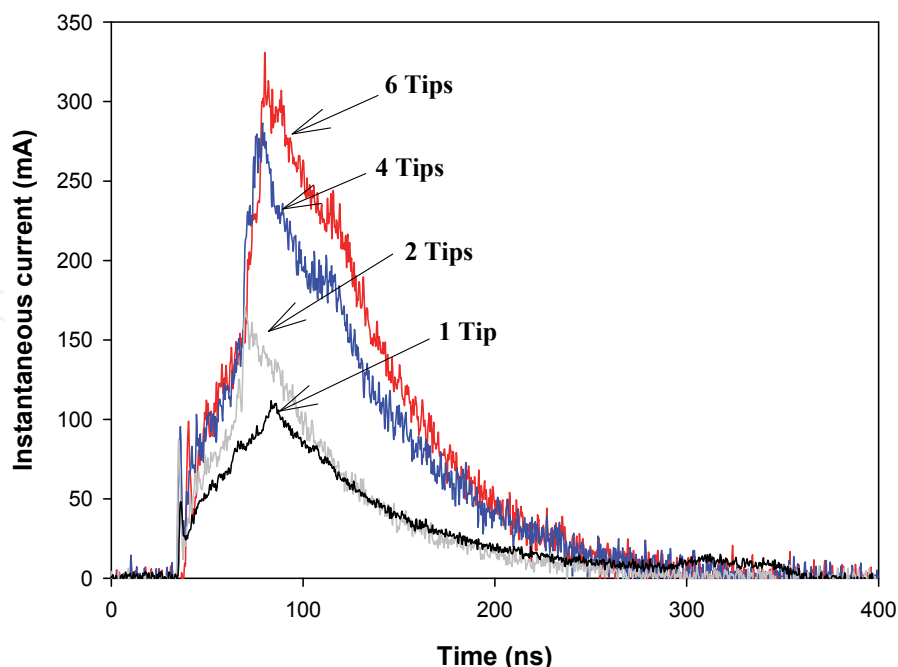


Fig. 4. Corona discharge current in the case of positive point to plane geometry as a function of time for different number of tips (when the number of tips is increased from 1 to 6, the peak current is increased only by a factor 3) from Abahazem et al 2011

2.4 Surface corona discharge

Surface discharges can be generated from many electrode configurations. A specific setup analyzed in our laboratory is displayed in Figs 5a, 5b and 5c (Gardou et al 2009). In such a case, both surface and volume discharges are present as displayed in Fig. 5c and also as already emphasized in the literature (see e.g. Allen et al 2004, Timatkov et al 2005).

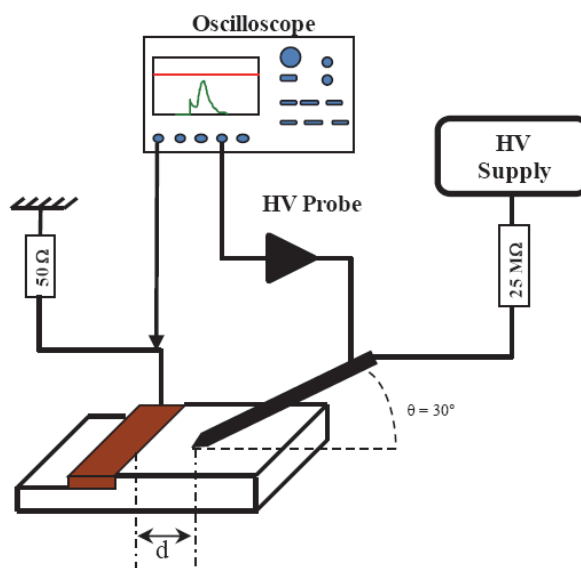


Fig. 5. a. Experimental setup of surface corona discharge with an inclination angle of 30° between the anodic needle and the dielectric surface (d being the gap separation between the tip of the needle and the cathode copper) from Merbahi et al 2011

However, the surface discharge is developed faster with a less energy consuming than its homologous in the volume because the main processes of the discharge propagation is the photo-emission from the dielectric in the case of the surface discharge (see e.g. Allen et al 2004) while it is the gas photo-ionization in the case of the volume discharge. As it is known the surface photo-emission requires less energy (2 or 3 eV) than the gas photo-ionization (e.g. 15.7 eV for N_2). This prefigures a better efficiency of active species production by the surface discharges in comparison to the classical corona volume discharge presented in sub section 2.3.

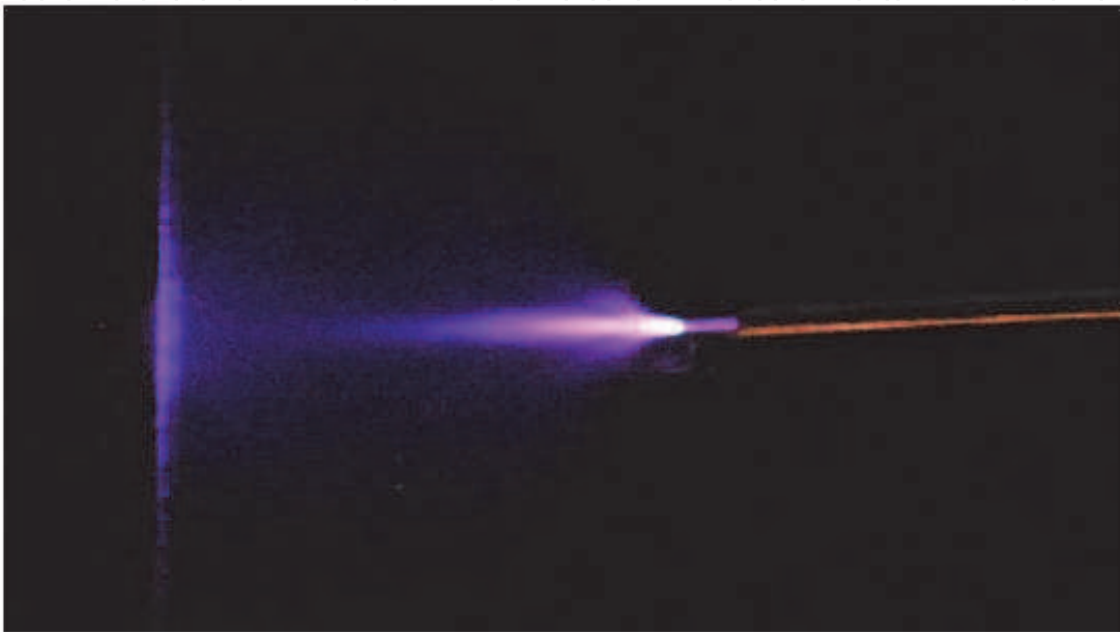


Fig. 5. b. Top view photograph of the corona surface discharge for radius tip $\rho = 50 \mu\text{m}$, $d = 13 \text{ mm}$, $U_0 = 11,5 \text{ kV}$, (cathode is on the left side, anodic point on the right side) in the case of PMMA insulator (from Gardou et al 2009).

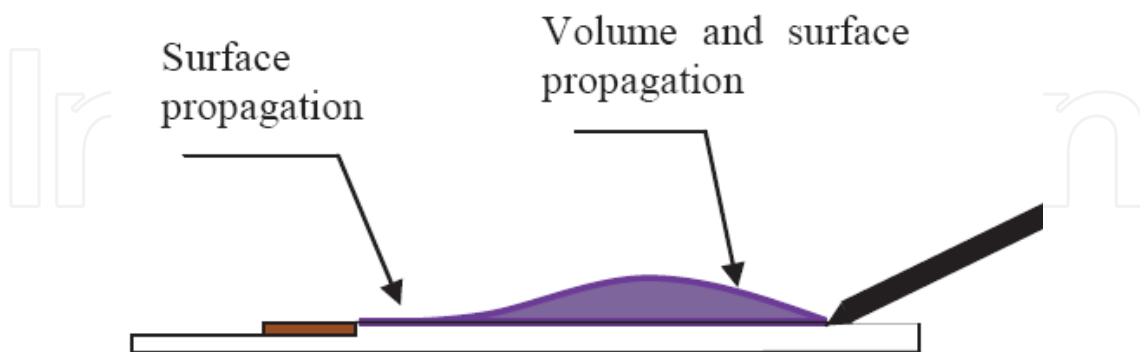


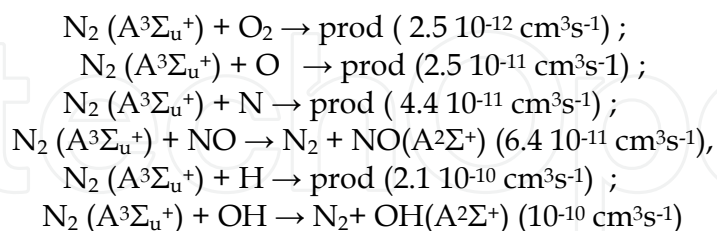
Fig. 5. c. Schematic cross section of the surface discharge in the case of PMMA insulator from (Gardou et al 2009).

2.5 Atmospheric pressure plasma jets

There are in the literature many devices developed to produce a low temperature plasma jet more particularly for biomedical uses as emphasized in for instance the review of Laroussi

2007. Plasma jet at atmospheric pressure allows a remote treatment very convenient for instance for in vivo treatment where it is dangerous to put the living tissue inside the zone of plasma generation. Plasma jet is also very interesting for biomaterial treatment more particularly when it is needed for instance to immerse the biomaterial in water for a treatment under hydrated form. There are many examples of plasma jet devices as for instance Plasma needle (Kieft 2005), plasma pencil (Laroussi 2005), plasma brush (Duan 2005) and other setups of plasma jets driven by various power supplies (DC or AC or RF and also micro-wave sources). Such plasma devices produce generally low temperature plasmas using various gas compositions and electrode configurations. In most cases, the plasma is launched outside the generation zone with the help of an external imposed continuous gas flow.

A specific low temperature plasma jet already used for the collagen treatment (Delaunay et al 2011) has been developed in our laboratory (see the patent of Merbahi et al 2011). The measured plasma temperature on the top of this specific plasma jet, that has a length of about 1 cm, does not exceed 27°C. This plasma jet shown in Fig. 6a is generated directly in the ambient air at atmospheric pressure and launched by itself without any external system of gas inlet feed that making it very easily transportable because there is neither gas bottle nor gas pumping (Merbahi et al 2011). It is generated by a specific corona discharge design giving a natural repetitive discharge current with a frequency of about 20 kHz under a high voltage power supply. Fig. 6b displays the instantaneous discharge current and the applied voltage that corresponds to a dissipated power of about 100 mW to generate the plasma jet. The UV-visible spectrum corresponding of the light emission of the top of the air plasma jet is displayed in Figs. 7ab. This shows the classical emission bands of Nitrogen such as the Second positive system (SPS) of $N_2(C^3\pi_u)_v \rightarrow N_2(B^3\pi_g)_{v'}$ from about 290 nm up to 440 nm corresponding to the different vibration states v and v' (for instance see the peaks at 315.93 nm, 337.13 nm, 357.69 nm, 375.54 nm and 380.49 nm). The first positive system (FPS) $N_2(B^3\pi_g) \rightarrow N_2(A^3\Sigma_u^+)$ which is apparent in the visible-near infrared (between about 600 nm up to 900 nm) range indicates this radiative way of the formation of metastable $N_2(A^3\Sigma_u^+)$ states. However, $N_2(A^3\Sigma_u^+)$ can be in turn quenched by the air species following several reactions given with their reaction rates (Herron, 1999):



The detection of the first negative system (FNS) of $N^+_2(B^2\Sigma_u^+) \rightarrow N+2(X^2\Sigma_g^+)$ around 390 nm is synonymous of high electron energies leading to the ion formation. However, concentration of positive nitrogen ion is probably very low due to the very small intensity of the FNS emission. There are also the oxygen emissions at for instance 759 nm coming from the band $O_2(b^1\Sigma_g^+ v=0) \rightarrow O_2(X^3\Sigma_g^- v=0)$ and the atomic line of the triplet state of O at 615 nm and 777.47 nm.

The $NO\gamma$ bands due to the emission of $NO(A^2\Sigma^+)_v \rightarrow NO(X^2\pi)_{v'}$ synonymous of dissociation of molecular nitrogen and oxygen leading to NO formation, (usually observable in classical corona discharges in quasi-pure N_2) are not observable in our plasma jet ambient air

between about 200 nm up to 290 nm due to the quenching of $\text{NO}\gamma$ bands by the molecular oxygen following the reaction: $\text{NO}(A^2\Sigma^+) + \text{O}_2 \rightarrow \text{NO}(X^2\Pi) + \text{O}_2$ ($10^{-10} \text{ cm}^3\text{s}^{-1}$) with a rate coefficient that is 1000 times higher than the $\text{NO}(A^2\Sigma^+)$ quenching by N_2 ($10^{-13} \text{ cm}^3\text{s}^{-1}$).

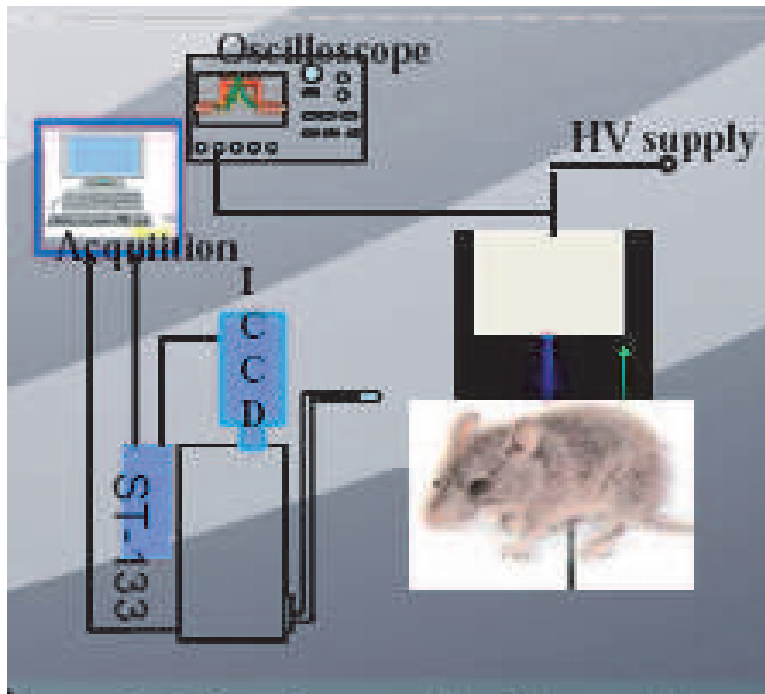


Fig. 6. a. Schematic view on the low temperature plasma jet generated in ambient and atmospheric pressure air.

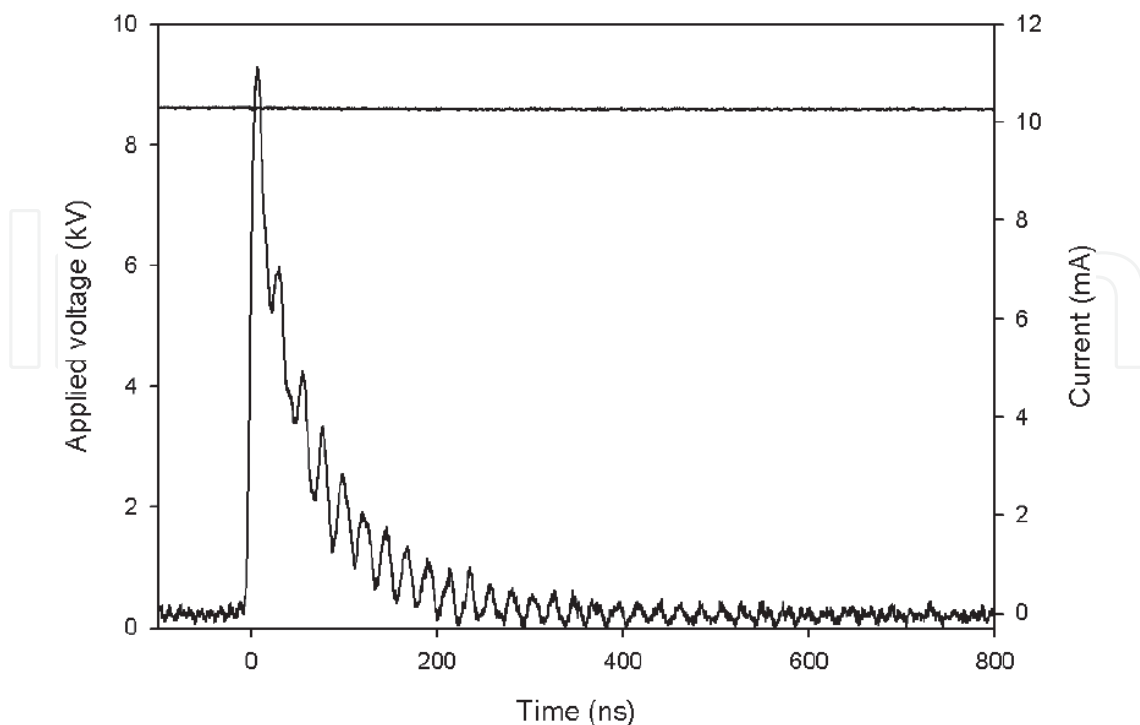


Fig. 6. b. Time evolution of the current of corona Discharge generating the plasma jet

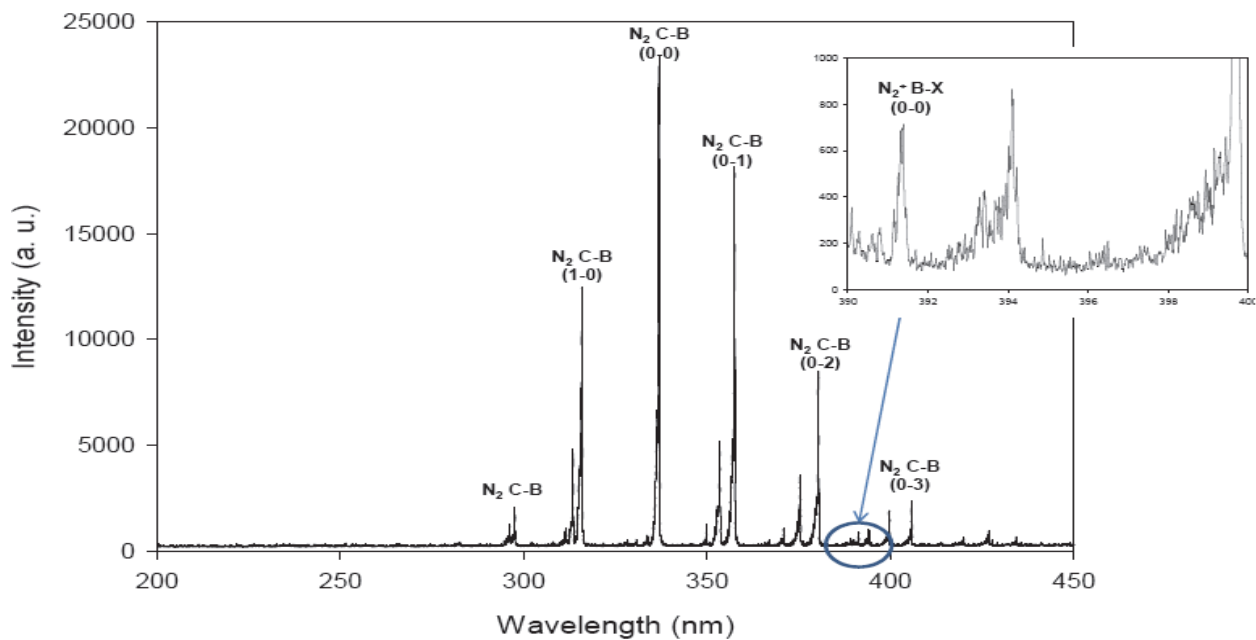


Fig. 7. a. UV-visible spectrum in the 200 nm-450 nm range collected in the top of the plasma jet.

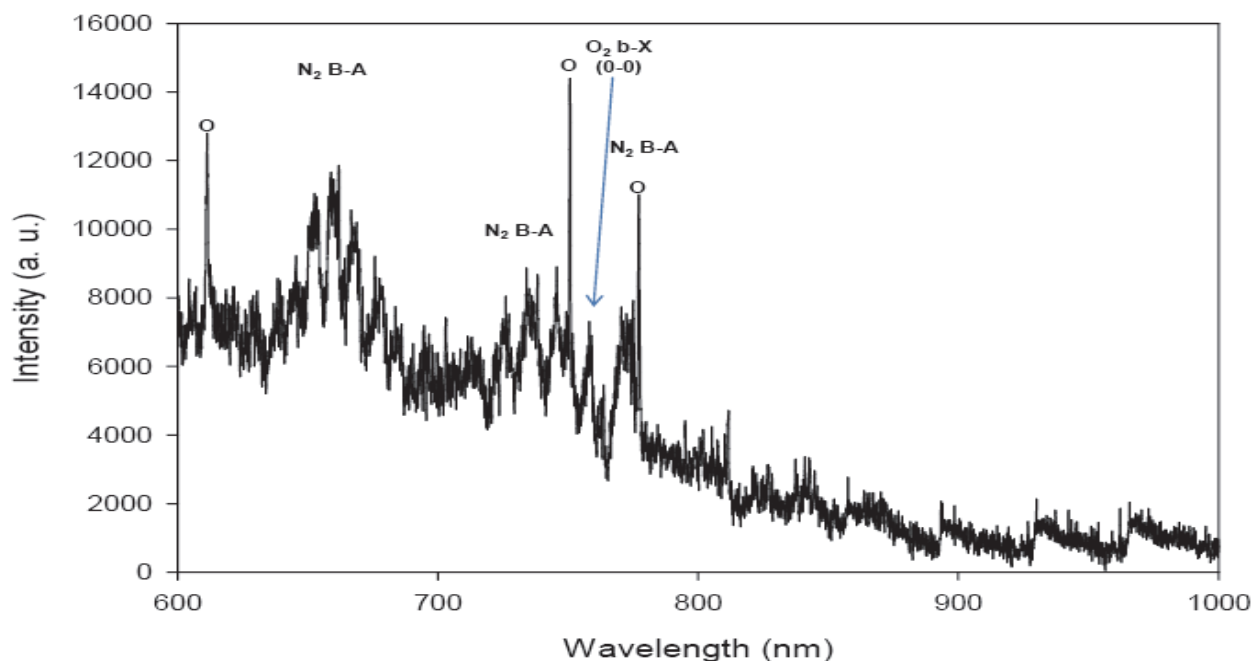


Fig. 7. b. UV-visible spectrum in the 600 nm-1000 nm range collected in the top of the plasma jet. NB: Between 450 nm up to 600 nm no significant emission has been observed.

The same remark is true for the $\text{OH}(A^2\Sigma^+) \rightarrow \text{OH}(X^2\Pi_{3/2})$ emission bands between about 300 nm up to 320 nm coming from the dissociation of water vapor impurities present in ambient air and not observed due to the quenching by molecular oxygen following the reaction: $\text{OH}(A^2\Sigma^+) + \text{O}_2 \rightarrow \text{OH}(X^2\Pi_{3/2}) + \text{O}_2$ with a rate coefficient that is roughly 10 times higher than the $\text{OH}(A^2\Sigma^+)$ quenching by N_2 .

In fact, the absence of emissions of $\text{NO}\gamma$ and $\text{OH}(A-X)$ can also be attributed to the quenching of metastable state N_2 ($A^3\Sigma_u^+$) that contributes in complement of electron collisions to the formation of the upper states of these two emission bands.

This overview on plasma jet spectrum means that the excited species present on the top of the jet are at least those involved by the emission bands from $\text{N}_2(C_3\Pi_u)_v$, $\text{N}_2(B_3\Pi_g)_v$, $\text{O}_2(b^1\Sigma_g^+ v=0)$, N_2 ($A^3\Sigma_u^+$) and O^* after electron collisions with O_2 and O . Due to the kinetics of formation of such excited species, it is obvious that we have also another metastable states of molecular nitrogen and oxygen, and also dissociation products of air such as the atomic nitrogen N and even the atomic hydrogen.

There are certainly another active species in the present ambient air plasma jet but they are not observable in the optical emission spectrum of Figs. 7ab due to mainly the quenching processes or to emission outside the present wavelength range (for instance specific atomic lines of nitrogen are in the VUV range less 200 nm and in the IR above 1000 nm).

3. Diagnostic tools for analysis and identification of active species

Dielectric barriers and Corona discharges generating non thermal plasmas in air or in other gas or gas mixtures at atmospheric pressure usually develop following streamer discharges having a thin filamentary structure (a few tens of μm wide) which propagates during a short time scale (a few tens of ns). A steady state glow discharge with a diffuse structure results at atmospheric pressure only from specific operating conditions (as e.g. a given frequency of the power supply and/or a chosen nature of the used dielectric and/or a specific gas composition: see e.g. Kelly-Wintenberg et al 1998).

Due to their short time and space scales, the streamer discharges are not easy to experimentally analyze and characterize in order to better know and to optimize these sources of active species. For these reasons, a large variety of plasma diagnostic tools covering optical, chemical and electrical methods are needed (see e.g. Creighton 1994, Ono et al 2005). These experimental tools can obviously be used for low temperature and non thermal plasmas generated at atmospheric pressure and also at reduced pressures. These experimental tools can be summarized as follows:

- OES (optical Emission spectroscopy) to detect radiative species with their temperature (see e.g. Ricard 2005, Zhao et al 2007)
- Absorption spectroscopy and FTIR (Fourier Transform Infra Red) spectral analysis to determine the species concentration (see e.g. Gorry et al 2007, Nair et al 2007)
- Laser spectroscopy such as LIF (Laser Induced Fluorescence) or TALIF (see e.g. Marchal et al 2010) to obtain the spatio-temporal evolution of the species concentration even at low proportion using CRDS (Cavity-Ring Down Spectroscopy)
- CARS (Coherent Anti-Stokes Raman Spectroscopy) for distribution of vibration molecules (see e.g. Oda et al 2006, Srivastava 2009, Stancu 2009)
- Image intensifier with CCD for spatial discharge structure analysis and streak camera for spatio-temporal analysis (see e.g. OCHKIN et al 1998, KEMPKENS et al 2000)
- Schlieren photography to visualize neutral gas variation and heat transfer from streamers to gas (see e.g. WINANDS et al 2006, Ohyama et al 2007)
- Mass spectroscopy to detect various ions and the stable radicals and neutrals, (see e.g. Held et al 1999, EBELING et al 2000)
- Gas chromatography to analyze stable oxide evolution before and after gas treatment (see e.g. CASANOVAS et al 1992)

- Electric measurements (voltage and current) to analysis electric behavior and to deduce energy discharge consumption (see e.g. Abahazem et al 2008, Merbahi et al 2008), etc.

4. Modeling tools for optimization of the source of active species

4.1 Overview on the different models

On the other hand, plasma modeling is also required to better understand and to optimize the formation and the behavior of our sources of active species. Plasma modeling and simulations which are very complementary tools to the experimental ones have generally the following main goals:

- a better understanding of basic phenomena and processes
- a good prediction of the plasma properties from validated models
- a reliable tool to research the optimal operating conditions for the plasma devices

Non thermal plasma modeling can concern a large variety of processes and phenomena contributing to the formation of active species and occurring during the discharge development and also during the post discharge stage. There are in the literature a lot of research works already devoted to modeling and simulations of the discharges or post discharge stages. A recent review on plasma modeling can be found in the paper of Lee et al 2011. However plasma modeling is generally undertaken without considering the strong coupling between the different models of for example streamer dynamics, gas dynamics, chemical kinetics and electrical circuit as it is summarized in the flowchart of Fig. 8. In fact when the streamer discharges develop and propagate, a streamer dynamics model is required to describe the spatio-temporal evolution of the streamer characteristics both in the streamer head and channel. The electric circuit model can give the input data for the streamer dynamics model. In the ionized channel left behind the streamer, there are a complex gas dynamics (gas pressure waves with gas density and temperature variations) influenced more particularly by the relaxation of excited species (metastable and vibration) which play the role of an energy tank: this is described by the gas dynamics model including the relaxation of excited states, the Joule effect corresponding to the energy transferred from electrons to the gas and the momentum transfer from ion towards neutral gas (ionic wind). A chemical kinetics model is also needed to describe the complex and various plasma chemistry involving the different active species whose the evolutions are initially affected by the streamer development and then are durably influenced by the local gas dynamics (gas temperature and density variations: see e.g. see e.g. Eichwald et al 2002). As it is shown in the flowchart of Fig. 8, each model is directly linked to the basic data models and therefore absolutely needs at its input the most reliable basic data in order to have the most reliable results.

In fact in our case of non equilibrium discharge at atmospheric pressure, the results of plasma models are first very sensitive to a lot of mathematical and numerical choices, approximations and parameters as for example:

- the good choice of the physical model (e.g. 1rst order i.e. continuity equation with momentum transfer conservation equation, second order i.e. up to energy conservation equation, ..) with its corresponding approximations (local field or local energy approximations, ...) (see e.g. Kanzari et al 1998)
- the good choice of the numerical scheme that have to take into account the strong coupling between transport, field and chemistry
- the numerical scheme is itself sensitive to the choice of criteria of stability and convergence, to boundary conditions, etc.

Results of plasma models are also very sensitive to the considered physical processes and phenomena and the associated basic data. If the aim of the plasma modeling is to have a quantitative description (not only a qualitative one) of our electrical discharge, it is necessary to obtain the various needed basic data with the best possible accuracy.

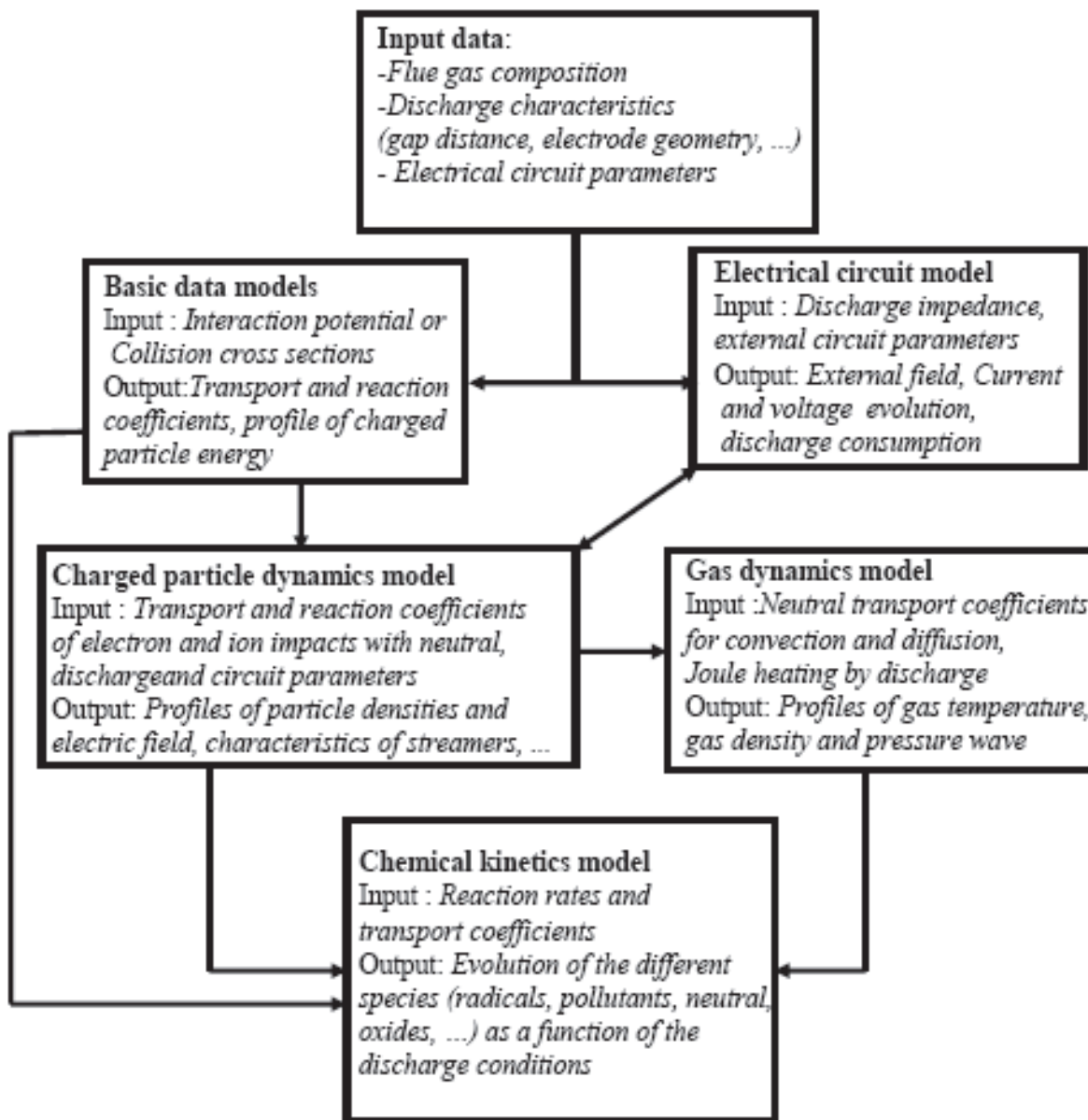


Fig. 8. Diagram showing the coupling between the different models which can be used to describe the electro-hydrodynamic and reaction kinetics phenomena and processes in the case of atmospheric pressure non thermal plasmas generated by corona or DBD discharges (Yousfi et al 2010)

4.2 Example of formation of specific active species in humid air discharge

In order to focus on the coupling of the different models previously evoked and the required corresponding basic data, let us consider the example of simplified reaction processes involved during the generation and the loss of radical O in humid air non

equilibrium discharge. As atomic oxygen can be a major active species in many biomedical applications, it is often necessary to quantify it very precisely as a function of the different discharge operating parameters (gas composition, power supply, design of electrodes, etc.) during the spatio-temporal discharge development.

Atomic oxygen in a humid air discharge is first affected by the dissociative electron impacts on O₂ and H₂O. This means that the knowledge of electron energy distribution (EEDF) or the electron transport and reaction have to be known as a function of the space charge electric field distribution.

Atomic oxygen density depends on ion-ion recombination processes, followed by electron detachment and ion conversions. This means the ion energy distribution (IEDF) or the ion transport and reaction has also to be known as a function of the space charge electric field distribution.

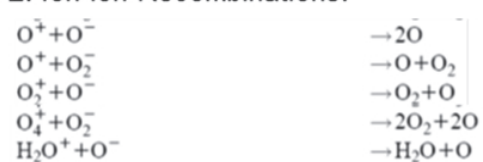
Furthermore, the production and loss of atomic oxygen depends on the reactions with metastable states and also on the reactions between radicals (N, OH, O, etc.) and background gas. This means that the distribution of excited species and radicals initially created by electron impacts have also to be precisely quantified as a function of the space charge electric field.

The evoked reactions involving electrons, ions, excited species and radical are summarized in table 1.

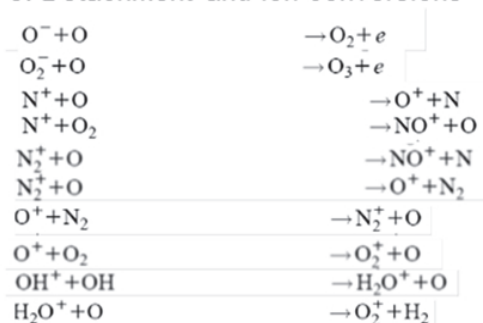
1. Electron impacts:



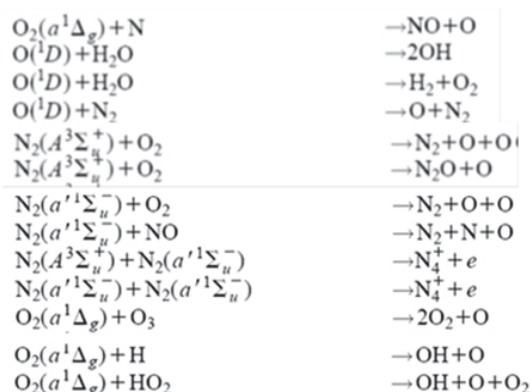
2. Ion-ion Recombinations:



3. Detachment and ion conversions



4. Reactions involving excited species:



5. Reactions involving radicals with background gas:

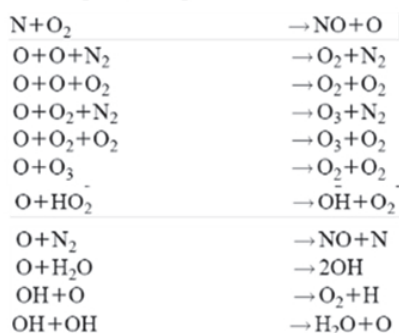


Table 1. Simplified set of reactions leading to production and loss of atomic oxygen in non equilibrium discharge in humid air

4.3 Electron basic data and energy distribution function

The input data needed to feed models are interaction potentials or collision cross sections, transport and reaction coefficients for electrons, ions and neutrals (i.e. radicals and excited species) interacting with the background gas. During the discharge development and propagation, the primary interactions are due to electrons which are the most energetic species. Thus electron-molecule collisions lead to the formation of the primary active species (dissociated, excited and ionized molecules). In order to accurately know the formation rate of the primary active species, electron energy distribution function (EEDF) have to be accurately known because it contains the complete information on the primary processes generating the studied non thermal plasmas. EEDF is obviously solution of Boltzmann equation of weakly ionized gas. The method recommended here to solve Boltzmann equation for the calculation of EEDF and then for electron transport and reaction coefficients has been described elsewhere (Yousfi 1996). It is based on the multi-term development of the distribution function (see in Fig. 9, the significant deviation between a Maxwell distribution, a two-term EEDF and a multi-term EEDF). The dominant interaction processes considered for electron Boltzmann equation are those between electrons and molecules in their ground state. However, the interactions between electrons and excited molecules have also to be considered as for instance superelastic collisions (molecular de-excitation) or stepwise ionization (ionization of a metastable state).

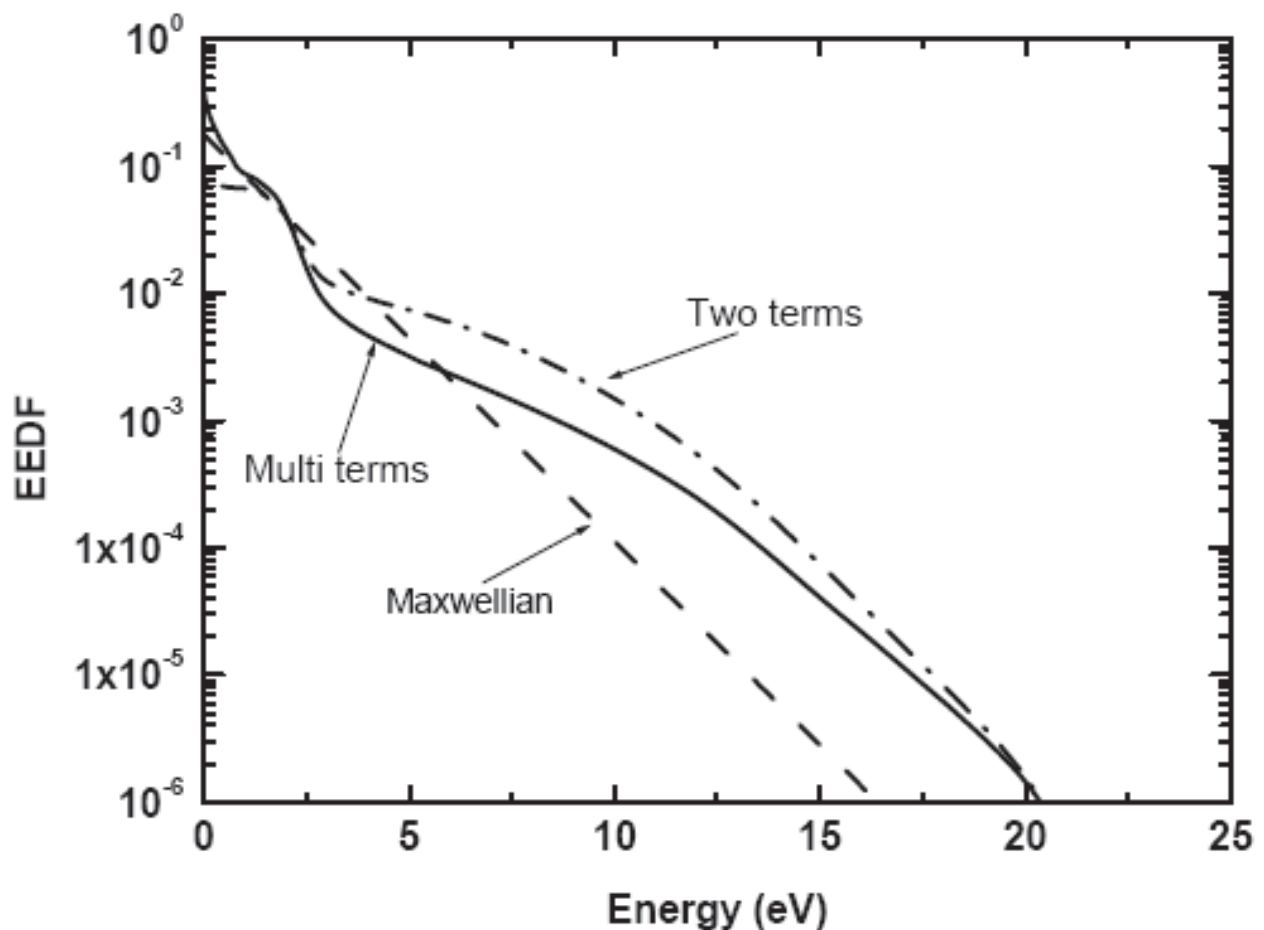


Fig. 9. EEDF as a function of electron energy in a humid gas mixture (76%N₂ 12%CO₂ 6%O₂ 6%H₂O) for an electric magnitude corresponding to the secondary streamer head or the primary streamer channel ($E/N=120$ Td)

Elastic collisions (using the Davidov operator for energy exchange and thermal agitation of molecules) and inelastic collisions (rotational, vibrational and electronic excitations, together with ionization and attachment) have also to be taken into account. Under these conditions, Boltzmann equation solution allows the calculation of the different electron transport and reaction coefficients measured in classical drift tube experiments. Then from EEDF of the different orders, we can obtain all the needed electron basic data such as excitation, ionization and attachment rate coefficients, and also electron drift velocity, transversal and longitudinal diffusion coefficients.

4.5 Necessity to use validated set of collision cross sections for distribution function computation

4.5.1 Electron-molecule collision cross sections

The use of an accurate EEDF is not sufficient, it is also necessary to use validated set of collision cross sections for the interactions of electrons with every molecules involved in the studied gas mixture (for example $e\text{-N}_2$, $e\text{-O}_2$, and $e\text{-H}_2\text{O}$ in the case of discharge in humid air at atmospheric pressure). The procedure of validation of every set of the electron-molecule collision cross sections (Yousfi et al 1996) is based on the known swarm unfolding technique summarized in the flowchart of fig 10. Figure 11 shows the adjustment done in the specific case of electron- H_2O set on the ionization cross section to better fit the recent measurements of the effective ionization coefficients based on time resolved Townsend experiment (de Urquijo et al 2007).

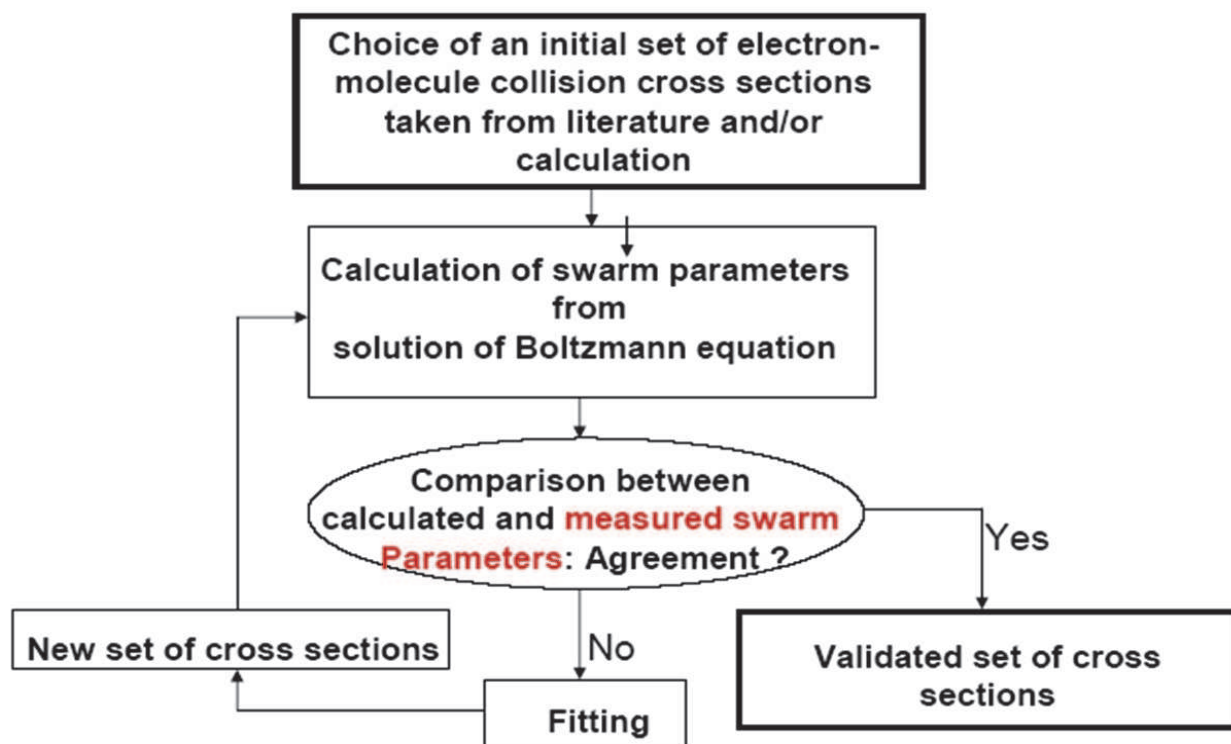


Fig. 10. Simplified flow chart of the fitting method of collision cross sections from Boltzmann equation solution (Yousfi 1996)

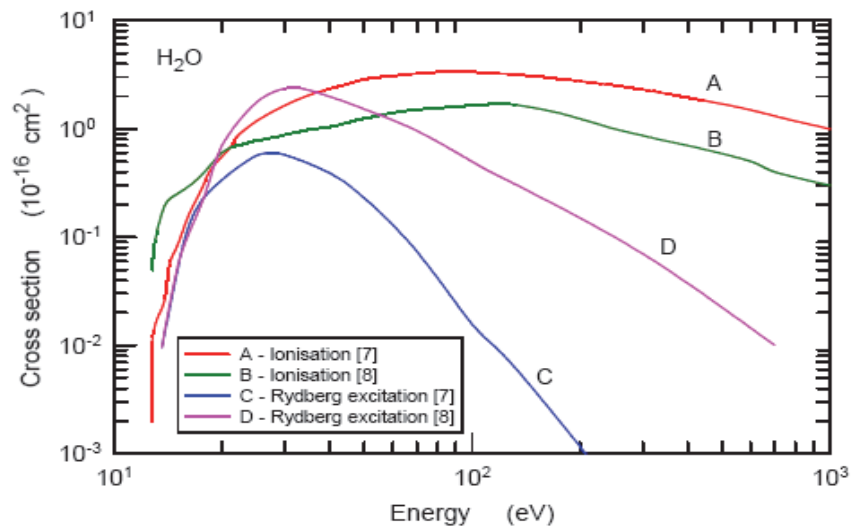


Fig. 11. a. Ionization and Rydberg excitation cross sections, taken from Yousfi 1996 (A and C [7]), and fitted by Yousfi et al 2010b (B and D [8]), from comparisons with recent Pulsed Townsend measurements of the effective ionization coefficients in H_2O and $\text{H}_2\text{O}-\text{N}_2$ mixtures (Ruiz et al 2010)

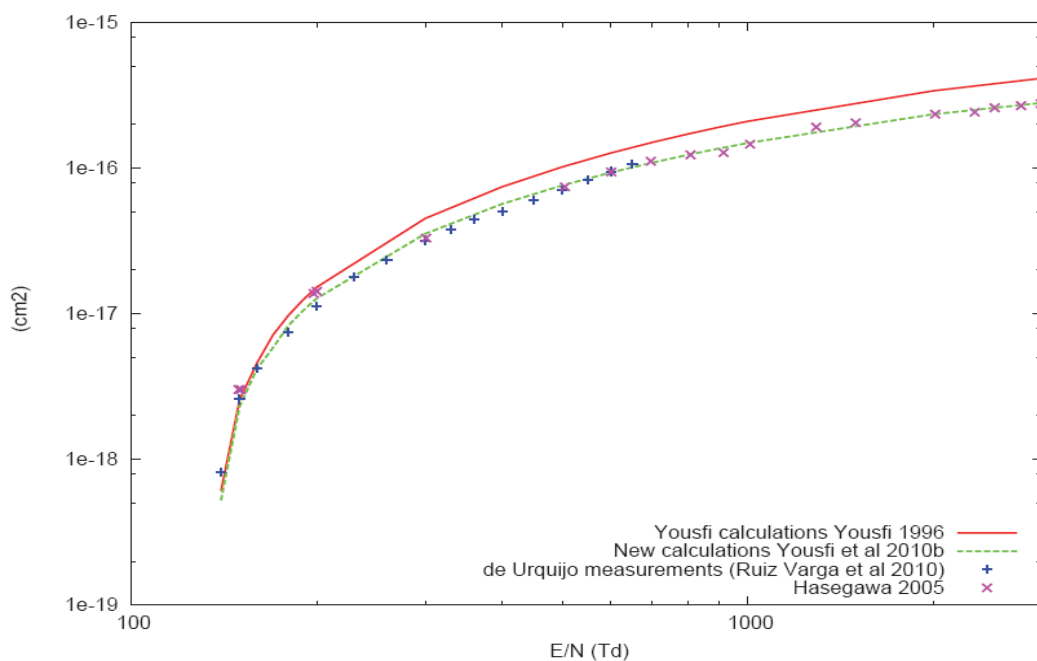


Fig. 11. b. Effective ionisation coefficient of electron in H_2O : comparison between calculation, measurements and literature data from Pulsed Townsend de Urquijo et al 2007 and Hasegawa et al 2005. The results, taken from Yousfi et al 2010b and Yousfi 1996, show that green curve is the best fit

4.5.2 Ion-molecule collision cross sections

It is also necessary to use validated sets of ion-gas collision cross sections because the discharge dynamics is also dependent on ion-gas collisions during the secondary streamer evolution and even in certain cases during the primary streamer evolution (Bekstein et al

2010). The involved ions in the discharge dynamics and in the ion chemistry in the case of humid air molecules are obviously the parent ions (N_2^+ or O_2^+ or H_2O^+) with the dissociated ions (N^+ or O^+ or O^-) and also some polyatomic ions (N_4^+ or O_4^+ or etc.) which can be created at atmospheric pressure following a three body reaction ($N_2^+ + 2N_2 \rightarrow N_4^+ + N_2$ or $O_2^+ + 2O_2 \rightarrow O_4^+ + O_2$) or two body reactions (interactions between two nitrogen or oxygen metastables) during the discharge dynamics and therefore can affect its development (Bekstein et al 2010). The transport and reaction coefficients of such ions, in the case of for instance a discharge in air, can be calculated from validated sets of collision cross sections for N_4^+ and O_4^+ ion in N_2 and in O_2 by using a JWKB method to calculate elastic momentum collision cross sections (Bekstein et al 2010) from a 12-6-4 or 8-6-4 core interaction potential $V(r)$ well adapted for diatomic and polyatomic ions.

$$V(r) = \frac{n\epsilon_m}{n(3+\gamma) - 12(1+\gamma)} \left[\frac{12}{n}(1+\gamma) \left(\frac{r_m - a}{r - a} \right)^n - 4\gamma \left(\frac{r_m - a}{r - a} \right)^6 - 3(1-\gamma) \left(\frac{r_m - a}{r - a} \right)^4 \right]$$

r is the intermolecular distance, ϵ_m the minimum energy of the potential, r_m the position of the minimum, n is the power of the repulsive part of the potential, γ is a strength factor and a is the shift between the mass center and the charge center (see Bekstein et al 2010 for details on the calculation and fitting on ion-molecule collision cross sections from $V(r)$). Fig. 12b shows an example of the obtained set of elastic and inelastic ion-molecule collision cross sections for systems O_4^+ / N_2 and O_4^+ / O_2 systems. Fig 12b shows the reduced mobility O_4^+ polyatomic ion in N_2 , pure O_2 and in air calculated from Monte Carlo method using the ion-molecule cross sections shown in Fig. 12a.

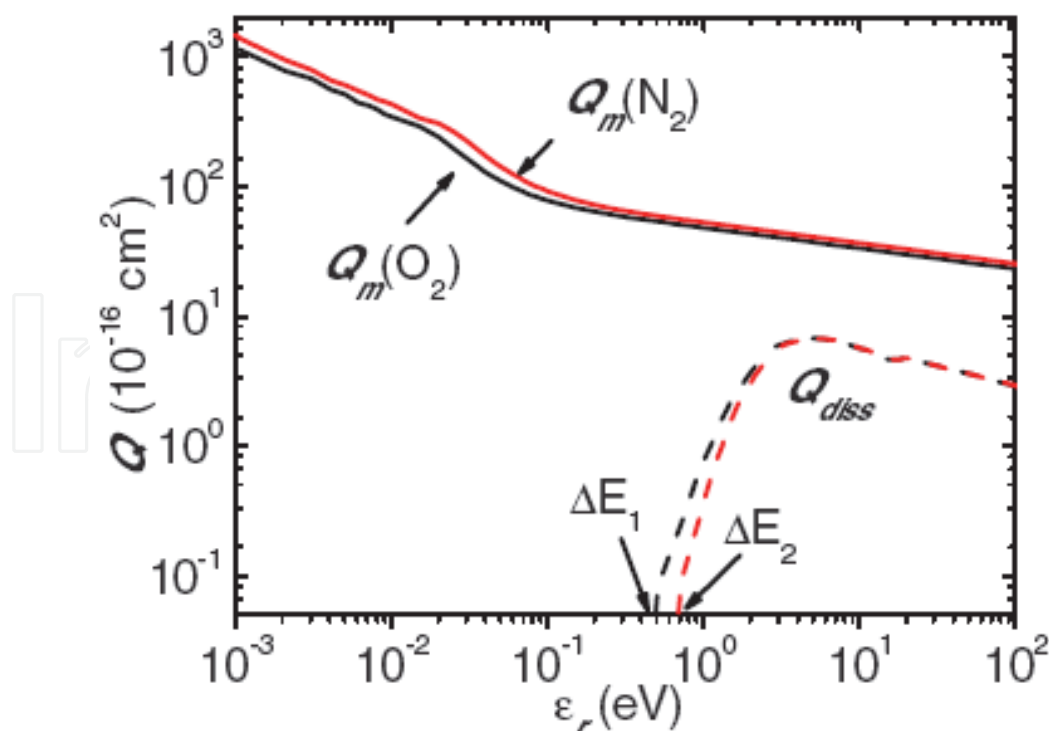


Fig. 12. a. O_4^+ / O_2 (black) and O_4^+ / N_2 (red lines) collision cross sections (taken from Bekstein et al 2010)

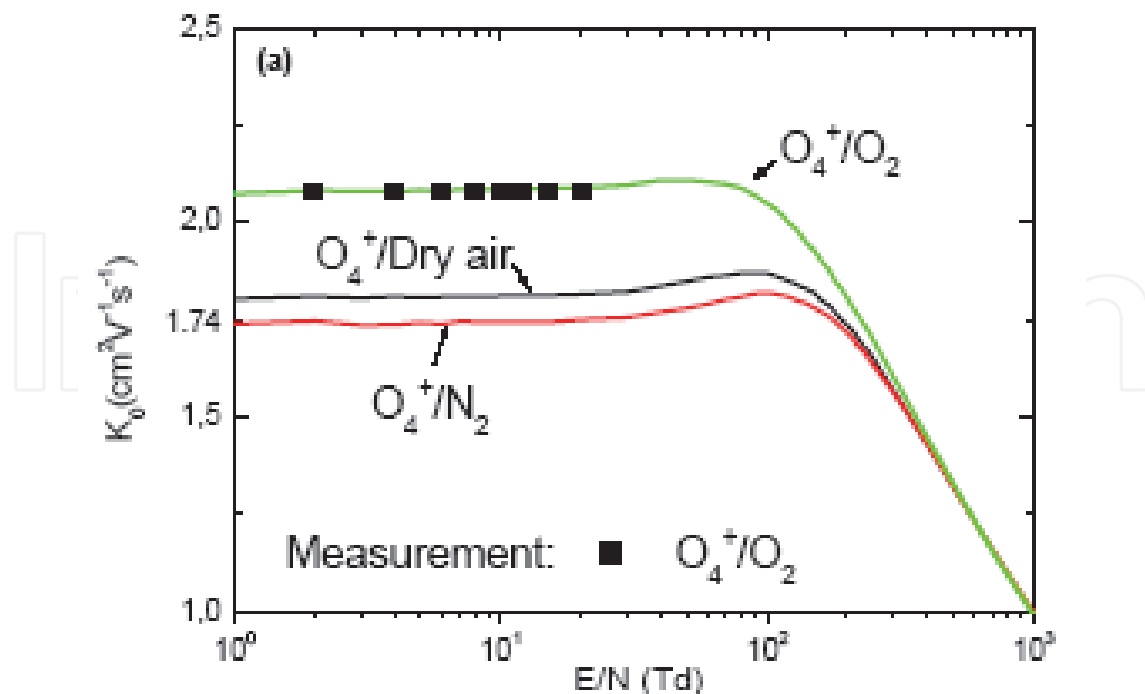


Fig. 12. b. Reduced mobility for O_4^+ ion in O_2 , N_2 and dry air (80% N_2 +20% O_2) calculated from Monte Carlo simulation (lines) and compared to available experimental data from Ellis et al 1976 (symbol) for O_4^+/O_2 system (taken from Bekstein et al 2010)

4.6 Example of the effect of the choice of ion basic data on streamer dynamics and active species production

The consideration of the previous basic data more particularly those of polyatomic ions (as for instance O_4^+) on the streamer dynamics modelling has obviously a significant effect on for instance the determination of the active species (as e.g. atomic radicals O and N) formation inside the ionized channel of the streamer during primary and secondary development. The effect of consideration of O_4^+ ion on streamer characteristics such as velocity, electric field propagation and charged particle densities is already discussed in Bekstein et al, 2010. In the following a focus is given specifically on the evolution of radicals like O and N atoms. Fig. 13 shows in the case of positive corona discharge between point-to-plane electrodes, the time evolution of the mean density (integrated over the whole gap distance from 0 up to 4mm) of radicals O and N produced during the propagation of primary streamer (from initial time up to 60 ns), the development of secondary streamer (up to about 75ns) and during the discharge relaxation (up to 200 ns). Radicals O and N can be (from such discharge modelling) quantified and localized during the discharge propagation and relaxation. We observe that these atomic active species which are very useful to identify and quantify for environmental and biomedical plasma applications, are mainly produced in the secondary streamer and discharge relaxation stages (due to the dissociative excitation and attachment of oxygen and nitrogen molecule). Such results are coherent with Ono & Oda 2004 measurements using UV Laser absorption spectroscopy. Furthermore we also observe, due to the better efficiency of O_2 dissociation, that O density is higher than N density despite the higher proportion of N_2 in dry air. Last, it is worth noting that the non consideration of the polyatomic ion overestimates, as expected, the densities of O and N

radicals; this emphasizes the importance of the polyatomic ion transport and reaction kinetics.

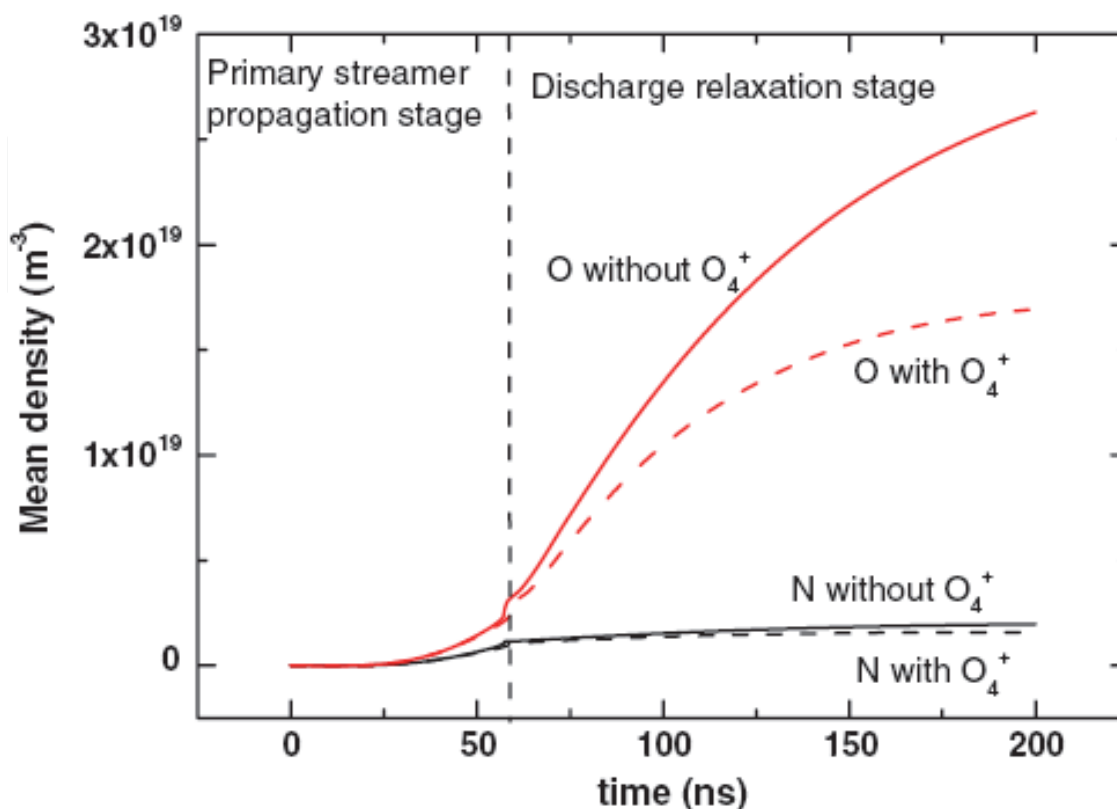


Fig. 13. Space integrated O and N radicals densities (integration over the whole space from 0 up to 4 mm) as a function of time in the case of a DC point-to-plane corona discharge in dry air at atmospheric pressure and a DC voltage of 5.3 kV (taken from Bekstein et al 2010)

5. Illustrative results

Two examples have been given in this section to illustrate the use of low temperature and non thermal plasma sources generating active species at both reduced and atmospheric pressures in the case of bacteria decontamination and bio material treatment.

5.1 Bacteria decontamination using sources of active species at reduced and atmospheric pressure

In the flowing afterglow discharge in pure N_2 at reduced pressure, the active species already identified (Villegier et al 2008) as the main decontamination vector are N atoms. The increase of the injected microwave power P_{MW} from 100 to 300 watts induces a 70% increase of the N concentration in the late afterglow reactor (Boudam et al 2007). Fig. 15 shows that such P_{MW} increase allows to obtain a 6 log decrease of the initial bacteria population after 30 min of exposure at room temperature. The microbiological protocole is described by Sarrette et al 2010. Fig. 15 also shows that in the case of the plasma jet in ambient air (CR-AP), a similar trend was obtained with a diminution of the distance d between the discharge and the exposed bacterial film.

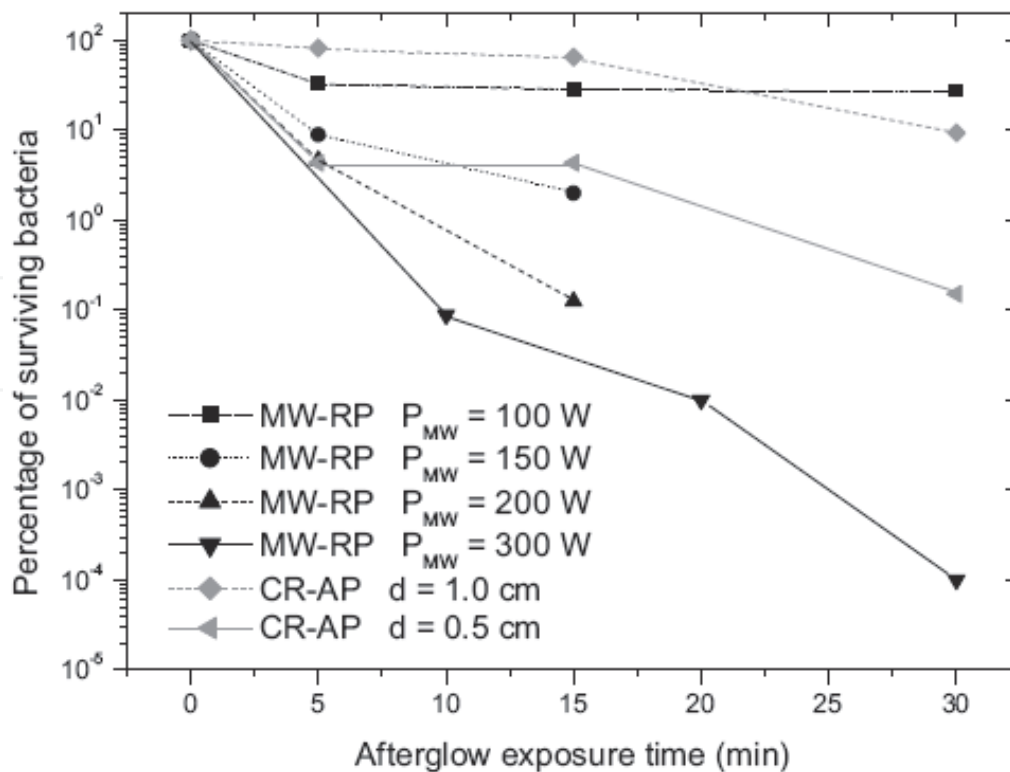


Fig. 14. Effect of increase of the concentration of the afterglow active species on the *E. coli* inactivation kinetics and effect of distance variation from the plasma jet. P_{MW} is the microwave power injected in reduced pressure discharge working in pure nitrogen (1 slm) at a 5 Torr (MR-RP) and d is the distance between the plasma jet in ambient air (CR-AP) and the exposed bacterial film (taken from Sarrette et al 2010).

This first example clearly emphasizes that the inactivation of *E. coli* bacteria can occur by using either a selective active species (atomic nitrogen) in the case of flowing afterglow discharge where the gas composition is well controlled or a mixture of active species (not clearly identified and quantified) in the case of the plasma jet in ambient air. This means that when the identification of the species really active in the biomedical processes, it is interesting to control totally the gas composition as in the case of flowing afterglow discharge at reduced pressure.

5.2 Collagen treatment using low temperature plasma jet in ambient air

Biomaterials based on collagen can be used as substitutes of the extracellular matrix for the realization of for instance skin substitutes. In order to develop new biophysical treatments to increase the durability and the stability of such biomaterials, plasma jet in ambient air has been tested (Delaunay et al 2010a and b) for the treatment of type I collagen which is the most abundant type in mammalian cells. Fig. 15 shows the Differential Scanning Calorimetry (DSC) graph of the control collagen sample (without plasma treatment) compared to the DSC graphs of collagen samples exposed to the ambient air plasma jet. Several exposure times are considered (5, 10 and 60 mn). A large endothermic phenomenon is observed for the different collagen samples between 50°C and 150°C. This can be attributed to the evaporation and vaporization of residual bound water in collagen (Samouillan et al 1999). A second endothermic event is observed above 200°C for all the

samples with a specific shape for each sample. This can be attributed to the denaturation of collagen. The later, distinct of degradation, implies that the rupture of peptide bonds, leads to the formation of an amorphous polymer. This more particularly means that the plasma treatment stabilizes the collagen structure because firstly there is a shift towards higher temperature range of the collagen denaturation and secondly there is a stiffening of the chains by a cross-linking action when compared to the control sample (collagen without plasma treatment). The thermal denaturation of collagen shows interesting information on the effects of plasma jet treatment on the triple helical structure and on the collagen stability. Further analyses are obviously needed to better quantify the plasma treatment on collagen by using the expertise of our colleagues of polymer physics (Samouillan et al 1999) and also biochemists in order to analyze the biomaterial functionalization.

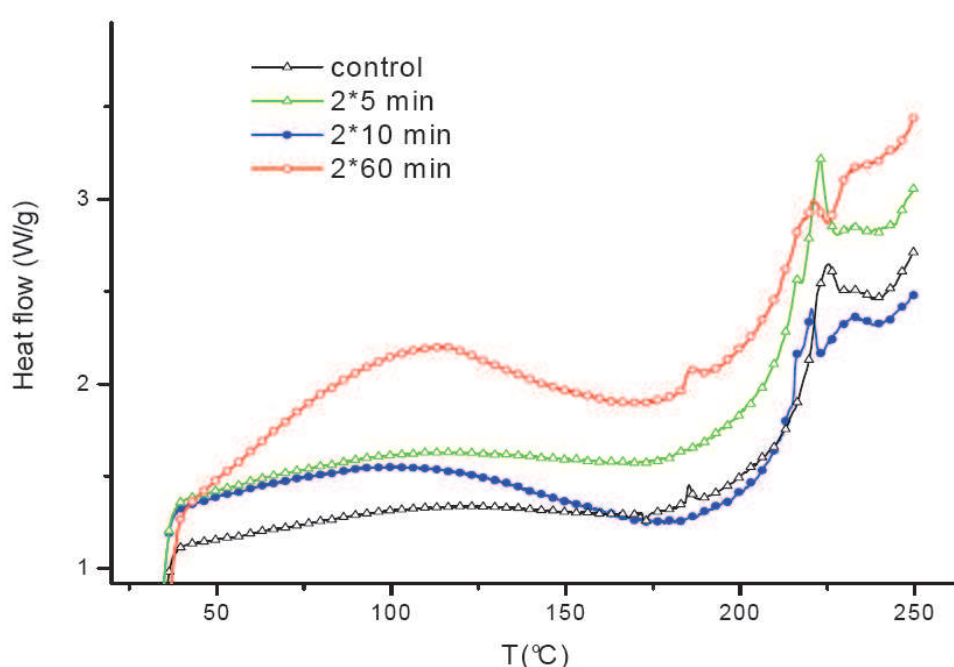


Fig. 15. Differential Scanning Calorimetry graphs of freeze dried collagen samples: comparison between control sample and treated (recto/verso) sample at different time exposures.

6. Conclusion

The optimizations of low temperature and non thermal plasmas have to be done in close collaboration between the plasma and biomedical researchers. For a given application concerning for instance decontamination or biomaterial functionalization or plasma medicine (as blood coagulation, wound healing, etc.) the role of the biomedical researcher is to finely analyze the effect of the different plasmas while the role of the plasma researcher is to provide a "tailored plasma" for the application. In fact, iterations are needed between plasma and biomedical researchers in order to identify and to select the plasma species (for instance a specific radical or a specific photon wavelength or a specific charged particle energy, etc.) really active in a given biomedical application.

In order to reach their goal, the plasma researchers need to use the experimental diagnostics and also modelling tools to better known and characterize the low temperature and non

thermal plasma as a function of the different operating parameters such as gas composition and pressure, shape and magnitude of power supply, geometry and design of the plasma reactor. To better understand and control the generation of active species more particularly in the case of streamer discharges at atmospheric pressure, further works are needed in both the experimental diagnostics and modeling of low temperature and non thermal plasmas. These works can concerns:

- the quantification of the spatio-temporal concentrations of radical and excited species (metastable and vibration) by using of advanced experiments such as LIF or TALIF and CRDS techniques adapted to the short transient regimes and filamentary discharge structure.
- the study of streamer branching phenomena by using fast imagery and 3D discharge dynamics modelling
- the physical understanding of photo-ionisation, photo-emission and secondary electrode processes (role of gas and surface on streamer development) that more particularly needs VUV and UV measurements of the short live excited species
- the model validation: further works are required on basic data for transport, chemistry and kinetics processes particularly in the case of polyatomic ions, excited species and radical interactions with gas and surface, etc.

Once developed and mastered the tools and the techniques of the characterization of the low temperature plasmas for their identification and quantification, the ultimate goal is to develop sources of active species fulfilling the following features:

1. production of selective active species well identified and well quantified,
2. very good efficiency of the production of the selected active species (i.e. high ratio between produced species and dissipated energy) and
3. last and not least the sources of active species have to be easily transportable, easily usable and without danger for the user.

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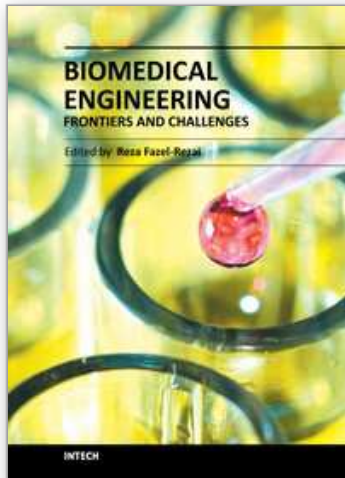
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In all different areas in biomedical engineering, the ultimate objectives in research and education are to improve the quality life, reduce the impact of disease on the everyday life of individuals, and provide an appropriate infrastructure to promote and enhance the interaction of biomedical engineering researchers. This book is prepared in two volumes to introduce recent advances in different areas of biomedical engineering such as biomaterials, cellular engineering, biomedical devices, nanotechnology, and biomechanics. It is hoped that both of the volumes will bring more awareness about the biomedical engineering field and help in completing or establishing new research areas in biomedical engineering.

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