Multi-dimensional simulation of a polluted gas flow stressed by a DC positive multi-pins corona discharge reactor

M. Meziane¹, O. Eichwald¹, J.P. Sarrette¹, O. Ducasse¹ and M. Yousfi¹,²

¹Université de Toulouse, UPS, INPT, LAPLACE (Laboratoire Plasma et Conversion d’Energie), 118 route de Narbonne, F-31062 Toulouse Cedex 9, France
²CNRS, LAPLACE, F-31062 Toulouse, France

Abstract—This paper is devoted to the complete multi-dimensional simulation of an Atmospheric Corona Discharge Reactor (ACDR) involving a maximum of 6 pins stressed by a DC high voltage and positioned 7mm above a grounded metallic plane. The ACDR is crossed by a lateral polluted air flow including 400ppm of NO. For the maximum time scale of 10ms considered in the present work, the simulation takes into account one hundred of successive discharge/post-discharge phases. The 2D and also the 3D simulations involve the radical formation and thermal exchange between the discharges and the background gas. The results show how the successive discharges activate the flow gas and how the induced turbulence phenomena affect the redistribution of the thermal energy and the chemical kinetics inside the ACDR. The 3D simulation results are presented for the first time and are compared with the 2D results during a time scale of 1ms.

Keywords—Positive DC atmospheric corona discharge reactor simulation, pollution control, flue gas chemical kinetic simulation

I. INTRODUCTION

Atmospheric Corona Discharge Reactors (ACDR) are efficient devices to generate active chemical species in proportion of some hundred to thousand of ppm. They are therefore interesting candidates in several low temperature plasma applications, like pollution control [1-6], ozone production [7,8], assisted combustion [9], biological surface decontamination [10] or biomedical field [11,12], where chemical reactivity is the main pathway of treatment without any significant increase of temperature. However, many complex electrohydrodynamics and thermal phenomena generated by the discharges can affect the efficiency of the ACDR i.e. the rate of formation or transformation of chemical species with respect to the electrical energy consumption and the nature of the chemical by-products. Therefore, as a complement to experimental studies, the complete and multi-dimensional simulation can be used to identify the main phenomena and reactions that influence the complex processes involved in an ACDR. However, in the field of corona micro-discharge study, several specificities considerably increase the difficulties such as for instance (i) the strong non stationary problem of alternating discharge and post-discharge phases with a repetition rate of tens of kilohertz, (ii) the large differences in the space and time scales between the very fast processes occurring during the discharge phase inside the small micro-discharge filaments and the lower ones covering a larger ACDR volume during every post-discharge phase or (iii) the judicious choice of a minimal set of both chemical reactions and species the most representative of the experimental observations while preserving reasonable computing times, among others.

Due to these specific difficulties, the complexity of ACDR simulation was progressively enhanced from uniform chemical kinetics [13-16] to simulation involving space non-uniformity in one or multi-dimensional domain and coupling one or several phenomena during the discharge and post-discharge phases [17-25]. Only recently, some works were devoted to the 2D simulation of successive discharge/post-discharge phases in a multi-pin-to-plan ACDR and for time scale extended up to some milliseconds [26, 27].

In the present paper, we present and analyse the results of a bi-dimensional and tri-dimensional simulation of an ACDR composed of 6 or 4 aligned pins crossed by a polluted dry air flow. We follow in details the spatio-temporal transformation of the NO pollutant until some milliseconds by coupling a maximum of 100 successive discharge/post-discharge phases with a repetition rate of 10kHz. The simulation involves the chemical kinetics and the energetic effects (including vibrational energy relaxation) of the electrical discharges on the neutral gas dynamics, temperature and reactivity.

Section II following this introduction is devoted to the description of the model and of the simulation conditions. Then section III describes and analyses the obtained results by showing more particularly how the successive discharges modify the air flow and its reactivity. The 3D results are also introduced for the first time and are compared with the 2D solutions.

II. METHODOLOGY AND SIMULATION CONDITIONS

It is obvious that the time and space scales between the discharge and the post-discharge phases are

Corresponding author: Olivier Eichwald
e-mail address: eichwald@laplace.univ-tlse.fr

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completely different. The micro-discharge generates plasma filaments of about $50\mu m$ in diameter during some hundreds of nanoseconds while post-discharge phases and the global chemical kinetic must be considered with decimetre scale and tens of milliseconds. Therefore, a complete simulation of both coupled phenomena for multi-pins reactor needs adaptive meshes from micrometer to centimetre scale and also adaptive time scale from picoseconds (in order to follow the nano-scale discharge phenomena) up to fraction of milliseconds. This means a large number of discrete spatial cells and a huge computing time. In order to overcome these difficulties, one can assume that the effects of the discharges on the background gas can be simulated by locally injecting (i.e. inside the specific micro-discharge volumes and only during the discharge phase) average source terms estimated from a complete discharge phase model.

Following the preceding remarks, the model used to simulate the ACDR involves the classical conservation equations of a reactive gas coupled with the conservation equation of excited vibrational energy and completed with specific source terms calculated from a multi-dimensional discharge phase model [22]. The set of equations are listed below:

\[
\frac{\partial \rho m_i}{\partial t} + \nabla \cdot (\rho v_i \vec{v}) + \nabla \cdot \vec{J}_i = S_i + \overline{S}_c \quad \forall i \tag{1}
\]

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0 \tag{2}
\]

\[
\frac{\partial \rho \vec{v}}{\partial t} + \nabla \cdot (\rho \vec{v} \vec{v}) = -\frac{\nabla P}{\rho} - \vec{v} \cdot \tau + \overline{S}_v \tag{3}
\]

\[
\frac{\partial \rho h}{\partial t} + \nabla \cdot (\rho h \vec{v}) = \nabla \cdot (k \nabla T) + \frac{\partial P}{\partial t} + \vec{v} \cdot \nabla P + \tau : \nabla \vec{v} \tag{4}
\]

\[
\frac{\partial \varepsilon_v}{\partial t} + \nabla \cdot (\varepsilon_v \vec{v}) = \overline{S}_c - \varepsilon_v \tag{5}
\]

Equations (1) to (5) are used to simulate the neutral gas behavior and to follow each neutral chemical species “$i$” (like N, O, O$_3$, NO$_2$, NO, ...) that are created inside the ACDR. $\rho$ is the mass density of the background neutral gas, $\vec{v}$ the gas velocity, $P$ the static pressure and $\tau$ the stress tensor. For each chemical species “$i$”, $m_i$ is the mass fraction, $\vec{J}_i$ the diffusive flux due to concentration and thermal gradients and $S_i$ the net rate of production per unit volume due to chemical reactions between neutral species. $h$ is the static enthalpy, $T$ the gas temperature, $k$ the thermal conductivity and $\varepsilon_v$ the vibrational energy.

The effects of the successive discharge phases are taken into account by specific sources terms. $\overline{S}_c$, simulates the creation of neutral active species (radicals and excited species) by electron or ion impacts with the main molecules of the background gas. $\overline{S}_v$ and $\overline{S}_m$ are the fractions of the total power density transferred during the discharge phase into thermal and vibrational energy. It is generally assumed that the translational, rotational and radiative electronic excitation energies relax quasi immediately into thermal form and that the vibrational energy stored during the discharge phases relaxes after a mean delay time $\tau_v$ of some tens of microseconds. This assumption explains the addition of equation (5) in the model. $\overline{S}_m$ is the total mean momentum transferred from charged species to the neutral background gas. All the previous source terms are estimated through a two dimensional (3D like) corona discharge model and are injected periodically in the thin discharge volumes with a repetition rate of 10kHz.

Fig. 1 and Fig. 2 show respectively the 2D and 3D geometry of the simulated ACDR. Both are composed by either 6 or 4 pins with a tip radius of 25$\mu$m. A grounded metallic plane is positioned at a distance $d=7mm$ below the pins and the inter-pin distance $e$ is constant and equal to 5mm. A DC positive high voltage of 7.2kV is applied on each pin and a stationary lateral atmospheric dry air flow polluted by 400ppm of NO crosses the ACDR from the right to the left hand side of the 2D or 3D domains. The discharge phases are characterized by the simultaneous propagation of vertical mono-filament discharges located between each pin and the grounded plane. The diameter of the mono-filament is equal to 50$\mu$m. In Fig. 2, the mono-filament discharges are represented by vertical thick lines around which the spatial grid definition is narrowed. The natural repetition frequency of the discharge phases is equal to 10kHz and each discharge phase lasts 150ns. The characteristics of each micro-discharge are supposed similar to an individual DC mono-pin-to-plane micro-discharge already studied elsewhere [22]. It means that the nature and the profile of the radicals and of the excited species injected in each micro-discharge are identical and that there is no spatial or mutual influence between two neighbouring micro-discharges.

The simulation involves a choice of 10 neutral chemical species (N, O, O$_3$, NO$_2$, NO, O$_2$, N$_2$, N$_2$ ($A$ $\Sigma_u^+$), N$_2$ ($a$ $\Sigma_u^-$) and O$_2$ (a$' \Delta g$)) reacting following 24 selected chemical reactions. The domain is discretized with square (2D) or cube (3D) structured meshes. The domains of injection of the discharge source terms have a size of 50$\mu$m$\times$7mm in the 2D case and 50$\mu$m$\times$50$\mu$m$\times$7mm in the 3D case. In these domains, located between each pin and the plane, we inject during 150ns specific source terms for the active species (namely : N$_2$($A$ $\Sigma_u^+$), N$_2$($a$ $\Sigma_u^-$), O$_2$(a$' \Delta g$)), N and O) and for energy that will simulate the micro-discharge effects.

The detailed explanations of the calculation of these specific source terms are given in reference [26]. For clarity, we remind here the main formulations. The discharge phase simulation gives all the source term profiles in a 3D mono-filamentary structure (2D cylindrical geometry with symmetry of revolution along
the discharge axis). The mean source terms of equation (1) to (5) are injected in very thin area so that it can be assumed only function of the $z$ coordinate which is aligned with the mono-filament discharge direction (see Figs. 1 and 2). Therefore, the mean source terms are expressed as follow:

$$ S(z) = \frac{1}{\pi r_d^2 l_d} \int_0^{l_d} \int_0^{2\pi} s(r, z, t) \, dr \, dt $$

(6)

$s(r, z, t)$ is the corresponding spatio-temporel source term calculate with the complete discharge phase model. $r$ is the radial coordinate (transversal to the discharge propagation direction in a 2D cylindrical geometry), $l_d$ is the characteristic discharge time (150ns in our case) and $r_d$ the characteristic discharge radius (50µm in our case). As an example, the mean source term $S_v$ in equation (5) is calculated through the discharge source term $s_v = f_v \dot{\rho} E$ where $f_v$ is the fraction of the total power density $\dot{\rho} E$ dissipated into vibrational excited energy.

Finally, the set of equations (1) to (5) is solved by the commercial FLUENT software [28].

![Design of the 2D simulated multi-pin-to-plane ACDR](image1)

**Fig. 1.** Design of the 2D simulated multi-pin-to-plane ACDR

![Design of the 3D simulated multi-pin-to-plane ACDR](image2)

**Fig. 2.** Design of the 3D simulated multi-pin-to-plane ACDR.

III. RESULTS AND DISCUSSION

A. 2D simulation results

Figs. 3, 4 and 5 show the spatio-temporal evolution of the gas temperature and of the ozone and NO concentration until 10ms. The cartographies at 0s show the initial conditions, i.e. a uniform profile of 300K for the gas temperature, no $O_3$ concentration and an uniform NO concentration of 400ppm ($9.7 \times 10^{15}$ m$^{-3}$). Time 0.1ms corresponds to the end of the first discharge/post-discharge phase, just before the injection of new source terms between the pin-to-plane gap. The results given at times 1ms and 10ms correspond respectively to 10 and 100 successive discharge/post-discharge phases. The cartographies are drawn just before a new discharge phase injection.

The temperature profile at 0.1ms (i.e. at the end of the first post-discharge phase) shows that the gas temperature rise is located only around the micro-discharge channels. Indeed, Fig. 3 shows that after 0.1ms (i.e. at the end of the first post-discharge phase and just before the second discharge phase), the gas temperature inside each filament reaches a maximum of 318K. The temperature remains higher near the stressed electrodes i.e. in the regions where the injected energy concentration is higher during the discharge phase. It should be noticed that at the end of the discharge phase (i.e. after 150ns), the gas temperature around each pin reaches about 1200°K [25]. During the post-discharge phase, this initially concentrated thermal energy is then transported in the gas flow and is distributed around each pin on a larger area by thermal diffusion. The increase of the temperature inside the discharge channels is also due to the relaxation of the energy stored into vibrational excitation state during the discharge phase. The relaxation time of this energy into thermal form is chosen equal to 50µs (i.e. 0.05ms) and its contribution to the gas temperature rise has been found equal to about 5K inside each micro-discharge volume. After 0.1ms, the decrease of the NO concentration inside the micro-discharge channels is mainly due to the reactions $O+NO+M \rightarrow NO_2+M$ and $O_3+NO \rightarrow NO_2+N_2$. Indeed, the discharge phase creates $O$ radicals inside each micro-discharge channel. The major part of them is rapidly consumed by the three body reaction $O+O_2+M \rightarrow O_3+M$ to form the stable $O_3$ molecules just after the discharge phase (see Fig. 5). It means that the oxidation reaction $O_3+NO \rightarrow NO_2+N_2$ is efficient in the ACDR volume where $O_3$ molecules are present while the three body reaction $O+NO+M \rightarrow NO_2+M$ is efficient only during the discharge phase and inside the micro-discharge channels.

At 1ms, 10 discharges phases have crossed the inter-electrode gap. The lateral air flow and the memory effect of the previous ten discharge phases lead to a plume formation of temperature, $O_3$ and NO concentrations. The plumes start from each pin, extend in the direction of the air flow and interact together. The results at 10ms show that the repetitive thermal shocks transform the laminar air flow into a turbulent flow. These thermal shocks
behave as successive obstacles for the initial laminar flow which disrupts into turbulent flow. Consequently, the chemical kinetic is no more only located inside the micro-discharge channels but is extended and distributed into the entire ACDR domain.

In fact, the Reynolds number in our simulation conditions can be estimated around $2 \times 10^4$ (with a velocity of $5\text{m.s}^{-1}$, a characteristic distance length of $1\text{cm}$ and a cinematic viscosity of air equal to $1,510^{-5}\text{m}^2\text{s}^{-1}$). In the case of a flow inside a cylindrical pipe, the transition between laminar and turbulent flow appears when the Reynolds number reach the threshold value of 2500. For a flow along an “infinite” plane, the turbulence transition appears for a typical Reynolds number of $5 \times 10^5$. Our situation can be considered in between as the flow is channelled by two infinite planes. Nevertheless, the Reynolds number is high enough so that small barriers in the initial flow (the successive thermal shock in our case) are able to generate turbulence.

One can notice that the cartographies of temperature, NO and O$_3$ concentration are very similar. In fact, the NO destruction is more pronounced in regions where the gas temperature is higher. Indeed, the oxidation reaction $\text{O}_3 + \text{NO} \rightarrow \text{NO}_2 + \text{N}_2$ is the predominant pathway to transform the NO pollutant in the ACDR volume. It means that the regions of lower concentration of NO correspond to the regions of higher concentration of O$_3$ (compare Fig. 4 and 5). Furthermore, the ozone and the thermal energy present the same behaviour. They are both injected inside the micro-discharge channel during the successive discharge phases and they accumulate in the turbulent regions of the ACDR where they are mixed in the air flow line. As a consequence, the cartographies of NO, O$_3$ and gas temperature present the same structure because of their great correlation.

B. 3D simulation results

Fig. 6 and 7 show the 3D cartographies of the gas temperature and of the NO concentration after 1ms of evolution i.e. after 10 discharge/post-discharge phases. The simulation domain corresponds to the one presented in Fig. 2 with 4 pins stressed by a DC high voltage. As in the case of the 2D simulation, the temperature and the NO density profiles follow the air flow lines and diffuse in the transversal directions. The temperature and the NO cartographies in the $(x,y)$ plan can be compared with the results obtained in the 2D simulation case. As in the 2D case, we observe the formation of plumes of temperature and concentration starting from each pin and extending in the direction of the gas flow. These plumes progressively interact together. At 1ms, the magnitudes of the variations of the gas temperature and the NO concentration are lower in the 3D case than in the 2D simulation case. In fact, starting from a similar temperature rise of 1200K near each pin at the end of a discharge phase, the thermal diffusion fluxes are involved in three dimensions i.e. through larger exchange surfaces. The initial accumulated thermal energy is therefore distributed in a larger volume than in the 2D simulation case. The NO molecules are mainly consumed in the oxidation reaction $\text{O}_3 + \text{NO} \rightarrow \text{NO}_2 + \text{N}_2$. However, the O$_3$ accumulation in the plumes is lower than in the 2D case because the diffusion of molecules takes place also in the two transversal directions $x$ and $y$. Furthermore, as soon as the NO concentration decreases, we observe a more important NO diffusion fluxes from
the high concentration region to the lower concentration region because of a more extended diffusion surface (3D rather 2D).

IV. CONCLUSION

The presented work shows the ability of the simulation to follow in detail the complex hydrodynamics and chemical kinetics that takes place in a multi-pins-to-plane ACDR. The simulation was extended until 10ms for 6 pins in the 2D simulation case and the results in the 3D simulation case were presented for 4 pins and until 1ms. The 2D simulation case has captured the turbulence formation starting from an initial laminar flow. The more realistic 3D simulation was presented for the first time. Even if the time scale was limited to 1ms for the 3D study, we found a good qualitative correspondence between the 2D and 3D results with however and as expected more pronounced diffusion phenomena. In fact, these phenomena take place in the volume of an ACDR and can not be efficiently simulated with a limited Cartesian 2D domain extension.

These preliminary results must be improved and confronted with experimental results obtained in very similar conditions. The study of larger domains and extended time scale would be performed with the use of parallel super computer.

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