DIRECT NUMERICAL SIMULATIONS
OF PLASMA-ASSISTED IGNITION IN
QUIESCENT AND TURBULENT FLOW CONDITIONS

M. Castela\textsuperscript{1,2}, B. Fiorina\textsuperscript{1,2}, A. Coussen\textsuperscript{1,2}, O. Gicquel\textsuperscript{1,2},
C. O. Laux\textsuperscript{1,2} and N. Darabiha\textsuperscript{1,2}

\textsuperscript{1}Ecole Centrale Paris, Grande Voie des Vignes, 92290 Chatenay-Malabry, France
\textsuperscript{2}CNRS, UPR 288, Laboratoire EM2C, Grande Voie des Vignes, 92290 Chatenay-Malabry, France

benoit.fiorina@ecp.fr

1 Introduction

Nanosecond Repetitively Pulsed (NRP) discharges are a promising technique to initiate and control combustion phenomena (Pilla et al. (2006); Kim et al. (2008); Adamovich et al. (2009); Starikovskiy and Aleksandrov (2013)). In such discharges, a part of the deposited energy increases the gas temperature whereas a substantial amount serves to excite high-energy electronic levels of molecules. Molecules are then dissociated via collisional quenching of excited states, for instance through reactions such as $\text{N}_2^* + \text{O}_2 \rightarrow \text{N}_2 + \text{O}_2$, where $\text{N}_2^*$ designates and excited electronic state of nitrogen, Popov (2001). This result was recently confirmed experimentally by Rusterholtz et al. (2013), who showed that up to 50% of molecular oxygen can be dissociated by NRP discharges through this process in the inter-electrode region. Experimental studies show that such high concentration of radicals has a beneficial effect on combustion processes (Pilla et al. (2006), Pancheshnyi et al. (2006), Ombrello et al. (2010), Sun et al. (2012)).

Some numerical experiments have recently been conducted in order to understand the influence of Nanosecond Repetitively Pulsed (NRP) discharges on the flame structure. Coupled detailed plasma and combustion kinetics simulations have been conducted for that purpose. As an example, Bak et al. (2012) have simulated plasma-assisted stabilization of a laminar premixed methane/air flame. Numerical investigation of the ignition of preheated H\textsubscript{2}/air mixtures excited by pulsed nanosecond discharges were performed by Yin et al. (2013) and Tholin et al. (2014).

In practical configurations, the problem complexity increases as plasma and combustion chemistry also interact with the turbulent flow. Radicals formed by the plasma discharges will initiate hydrocarbons reactions and turbulence will connect and diffuse these radicals leading to a decrease of their local concentration and, therefore, their efficiency on initiating combustion processes. Consequently, during ignition, there is a competition between NRP discharges and turbulence characteristics on the combustion enhancement.

The aim of the present study is to numerically investigate these interactions. A direct numerical simulation (DNS) taking into account both detailed plasma chemistry and detailed combustion chemical kinetics still remains out of reach due to high CPU costs. In the present work, in order to focus on the effects of both temperature and radicals on the ignition process, we model the plasma discharge physics and kinetics based on experimental and numerical results. Nonetheless, a detailed combustion kinetic mechanism is considered in the DNS to capture ignition phenomena. We will analyze the first phase of the ignition process during which the initial flame kernel is formed in both quiescent and turbulent flow conditions.

2 Characteristics of NRP discharges

NRP discharges are characterized by high-voltage pulses (from 1 to 10 kV) that last for few nanoseconds and are repeated with frequencies of the order of tens of kHz. As a consequence of this applied voltage, a strong electric field is formed causing an increase of electron kinetic energy. This kinetic energy is then transferred through a cascade of collisions to the surrounding heavy species, giving rise to excited particles, ions and atoms. The distribution of the discharge energy over the various excited states of molecules depends on the electric field created by the discharge and on the local gas composition and temperature. The species in high energetic modes (vibrational and electronic modes) will eventually collide with the surrounding heavy particles and transfer their stored energy into the dissociation of molecules and/or gas heating.

In contrast with combustion processes where gas sensible energy is described by a single quantity, defined by a single local gas temperature, NRP discharges produce internally excited molecules over ro-
tational, vibrational and electronic modes that are usually not in equilibrium with the translation mode.

Modelling NRP discharges along with combustion processes requires, therefore, coupling non-equilibrium with equilibrium formulation. For this, the present model looks at two major NRP discharges characteristics observed experimentally and numerically. The first is the distribution of the discharge energy over two major groups of energy levels (electronic and vibrational levels) and the second is the relaxation time of these energetic levels towards equilibrium with translation modes. As we discuss below, electronic-translational equilibrium is approximately reached within the discharge characteristic time, whereas vibrational-translational equilibrium takes longer.

2.1 – Ultra-fast phenomena: relaxation of electronic states of molecules with a characteristic time of 20 to 50 ns.

Experimental studies presented by Rusterholtz et al. (2013) and Stancu et al. (2010) show that, for NRP discharges in air at atmospheric pressure and initial temperature of 1000 K, N₂ molecules are excited to N₂(B) and N₂(C) electronic states. It is also shown that the population of these N₂ excited states may result in the dissociation of around 50% of molecular oxygen and the increase of 900 K of the gas temperature, inside the discharge channel. By simple energy balance computation, Rusterholtz et al. (2013) have concluded that about 35% of the discharge energy is transferred into O₂ dissociation, while about 10 to 30% is associated to the fast increase of gas temperature inside the discharge channel. In conclusion, the discharge energy stored in electronic states of N₂ molecules via electron impact produces oxygen dissociation and heating in about 20 ns, through dissociative quenching reactions with molecular oxygen:

\[ N₂(X) + e^- \rightarrow N₂(A,B,C,d,...) + e^- \]

\[ N₂(A,B,C,d,...) + O₂ \rightarrow N₂(X) + 2O + heat \]  (1)

This two-step mechanism, which is an extension of the one proposed by Popov (2001) is known as the ultra-fast gas heating mechanism. Several other experimental studies, Bak et al. (2012), Adamovich et al. (2009), have also corroborated this rapid formation of atomic oxygen and temperature increase during the discharge characteristic time (of the order of few tens of nanoseconds).

2.2 – Slow phenomena: relaxation of vibrational states of molecules with a characteristic time from 1 µs to milliseconds

The remaining portion of the discharge energy is transferred to the vibrational levels of N₂ molecules (Popov (2001), Flitti and Pancheshnyi (2009), Aleksandrov et al. (2010)). The vibrational states of N₂ molecules may last for a long period of time in the mixture, according to Millikan and White (1963) before an equilibrium is reached between vibrational and translational modes. Millikan and White (1963) experimentally studied the characteristic vibrational-translational (VT) relaxation time of N₂ molecules for a wide range of gas temperatures and different concentrations of atomic and molecular oxygen in the gas mixture. The authors derived an empirical correlation that allows the computation of this characteristic time as a function of the local gas composition and the mixture temperature:

\[ \tau_{VT}^k = c / p_k \cdot \exp[α_k \cdot (T^{-1/3} - b_k)] - 18.42 \]  (2)

where \( \tau_{VT}^k \) is the vibrational-translational relaxation time of N₂ molecules by the \( k \)th collisional species partner (O₂, O and N₂), \( c_k \) is the partial pressure of the \( k \)th species and \( α_k \) and \( b_k \) are experimental constants depending on the \( k \)th species. The results have shown that the rate of energy exchange between vibrational and translational states (heating process) is highly sensitive to the partial pressure of atomic oxygen in the mixture.

3 Reactive flow governing equations for Ignition assisted by NRP discharges

3.1 Governing equations:

The governing equations describing a multi-component reactive flow subject to NRP discharges are:

\[ \frac{\partial \rho}{\partial t} + \frac{\partial (\rho u_i)}{\partial x_i} = 0 \]  (3)

\[ \frac{\partial (\rho u_i)}{\partial t} + \frac{\partial (\rho u_i u_j)}{\partial x_j} = \frac{\partial p}{\partial x_i} + \frac{\partial (\tau_{ij})}{\partial x_j} \]  (4)

\[ \frac{\partial (\rho \omega_i)}{\partial t} + \frac{\partial (\rho u_i \omega_j)}{\partial x_j} = \frac{\partial (\rho \omega_i)}{\partial x_i} + \frac{\partial (\tau_{ij})}{\partial x_j} + S_{\omega} + R_{\omega} \]  (5)

\[ \frac{\partial (\rho \sigma_{ij})}{\partial t} + \frac{\partial (\rho u_i \sigma_{ij})}{\partial x_j} = \frac{\partial (\rho \sigma_{ij})}{\partial x_i} + \frac{\partial (\tau_{ij})}{\partial x_j} + S_{\sigma} - R_{\sigma} \]  (6)

\[ \frac{\partial (\rho Y_k)}{\partial t} + \frac{\partial (\rho (u_i + V_{k,i}) Y_k)}{\partial x_i} = W_k \omega_i + Y_k \omega_k \]  (7)

where \( \rho \) is the density, \( u_i \) the velocity component in \( x_i \) spatial direction, \( p \) the pressure. \( \tau_{ij} \) refers to the viscous tensor, and \( \sigma_{ij} = \tau_{ij} - p \delta_{ij} \). In Eq. (5), \( \omega_i \) is the total energy which includes sensible (translational), chemical and kinetic contributions (Poinson and Veynante (2011)). In Eq. (6), \( \sigma_{ij} \) is the sensible vibrational energy. \( Y_k \) is the \( k \)th species mass fraction. The diffusion velocity of the \( k \)th species, \( V_{k,i} \), is computed assuming complex transport phenomena.
as in Coussen et al. (2012). \( W_k \) represents the molar production rate of the \( k \)th species due to detailed combustion chemistry. The source terms \( S_{\mathrm{et}} \) and \( S_{\mathrm{sp}} \), in energy equations Eq. (5) and Eq. (6) respectively, correspond to the external source of energy due to the electric discharge and \( \alpha \omega^v \) is the kth species molar production rate due to the electric discharge. The first term on the RHS of Eq. (6) refers to the diffusion of the vibrational energy, where the diffusion coefficient of nitrogen, \( D = D_{N2} \), is considered. \( \mathcal{R}_v^c \) is the coupling source term for the vibrational-translational energy exchange. The models for \( S_{\mathrm{et}} \) \( S_{\mathrm{sp}} \), \( \mathcal{R}_v^c \) and \( \alpha \) are discussed next.

### 3.2 – NRP discharge model:

Based on the literature results presented in section 2, a simplified model of NRP discharge-assisted combustion is constructed. Figure 1 shows a schematic representation of the model’s assumptions. The deposited discharge energy is assumed to be distributed within 50 nanoseconds into three components (ultra-fast heating, ultra-fast \( O_2 \) dissociation and vibrational excitation) as described in the following paragraphs.

#### 3.2.1 - Plasma energy model

During the ultra-fast phenomena, a fraction \( \alpha \) of the discharge energy is deposited into the mixture by the ultra-fast phenomena within a characteristic time of 50 ns (Fig. 1). Assuming a linear deposition of this energy, the source term in Eq. (5) is modeled as follows:

\[
S_{\mathrm{et}}^\alpha(t_{\text{per}}, x) = \begin{cases} \alpha \cdot \alpha_{\text{pulse}} \cdot f(x, t_{\text{per}}) / \tau_d & \text{if } t_{\text{per}} \leq \tau_d \\ 0 & \text{if } t_{\text{per}} > \tau_d \end{cases}
\]  

where \( \alpha_{\text{pulse}} \) refers to the discharge energy density (J/m³) and \( t_{\text{per}} \) refers to the time in the pulse reference. The function \( f(x, t_{\text{per}}) \) spatially defines where the plasma discharge occurs and is modeled as follows:

\[
f(x, y) = \text{erfc}\left(\frac{r(x, y)}{a}\right)^{1/e}
\]

The remaining portion (1-\( \alpha \)) is assumed to be transferred to the vibrational states of molecules, thus increasing the mixture vibrational energy, \( e_{\text{vib}} \) (Fig. 1 – Ultra-fast phenomena). This vibrational energy will eventually relax into heating but within a longer period of time (Fig. 1 – Slow phenomena). In turbulent conditions and strong convective flow field, there is a transport of this vibrational energy before it heats up the mixture and thus, the relaxation of this energy may occur away from the discharge channel. Therefore, a vibrational energy transport equation is introduced in Eq. (6) to transport this remaining energy, pulse after pulse. The model combines the energy of all possible vibrational states of molecules into a single term, \( e_{\text{vib}} \). The source term for this equation, \( S_{\text{sp}}^\alpha \), is given by:

\[
S_{\text{sp}}^\alpha(t_{\text{per}}, x) = \begin{cases} (1 - \alpha) \cdot \alpha_{\text{pulse}} \cdot f(x, t_{\text{per}}) / \tau_d & \text{if } t_{\text{per}} \leq \tau_d \\ 0 & \text{if } t_{\text{per}} > \tau_d \end{cases}
\]

The sink term, \( \mathcal{R}_v^c \) of Eq. (6) models the relaxation rate of vibrational energy into gas heating, and thus, it appears as a source term in Eq. (5). To model \( \mathcal{R}_v^c \), the harmonic oscillator approach of Landau-Teller is assumed:

\[
\mathcal{R}_v^c = \frac{\rho \cdot e_{\text{vib}}^\alpha(T) / \tau_{\text{re}}}{\tau_{\text{re}}}
\]

in which the equilibrium value of the vibrational energy at a given mixture temperature, \( e_{\text{vib}}^\alpha(T) \), is defined as:

\[
e_{\text{vib}}^\alpha(T) = \frac{\rho \cdot \Theta_{\text{vib}}}{e_{\text{vib}}^{\text{eq}} - 1} \text{ with } \Theta_{\text{vib}} = 3396 \text{ K}
\]

where \( \Theta_{\text{vib}} \) is the vibrational temperature corresponding to the first quantum vibrational state and \( r = R / W_{N2} \), with \( R \) the gas constant and \( W_{N2} \) the nitrogen
molar mass. The characteristic vibrational-translational relaxation time, \( \tau_{VT} \), is computed as a function of the \( \tau_{VT}^k \), for \( k = O, O_2 \) and \( N_2 \) species, given by the experimental correlation of Millikan and White, given in Eq. (2):

\[
\tau_{VT} = \left( \frac{1}{\tau_{VT}^O} + \frac{1}{\tau_{VT}^{O2}} + \frac{1}{\tau_{VT}^{N2}} \right)^{-1}
\]  

(13)

3.2.2 - Plasma model

A fraction of the total discharge energy serves to dissociate \( O_2 \) into \( O \) atoms by the ultra-fas mechanism (see Fig. 1). Therefore, the molar production rate of \( O \) and \( O_2 \) molecules by plasma discharges is modeled as follows:

\[
\omega^O = \eta_{diss} \cdot \sigma_{\text{mole}} \frac{Y_{O_2}}{\tau_d} \frac{1}{h_e^o}
\]

\[
\omega^{O_2} = \frac{W_O}{W_{O_2}} \omega^O
\]

\[
\omega^k = 0, \quad \text{if} \quad k = O, O_2
\]

(14)

(15)

(16)

where \( \eta_{diss} \) is taken equal to 35\% following Rusterholz et al. (2013) and \( h_e^o \) stands for molar enthalpy of atomic oxygen. \( Y_{O_2} \) refers to the \( O_2 \) mass fraction and the ratio \( Y_{O_2}/Y_{O_2}^\text{air} \), ensures that the dissociation rate of \( O_2 \) tends to zero when all dioxygen is consumed. \( W_O \) and \( W_{O_2} \) stand for the atomic and molecular oxygen molar masses.

4 Numerical experiment configuration and setup

The present model is implemented in a structured Direct Numerical Simulation solver dedicated to compressible reactive flow simulations with detailed chemistry (Coussen et al. 2012). The spatial derivatives are computed with a \( 4^\text{th} \) order centered finite-difference scheme. An \( 8^\text{th} \) order filtering scheme is used for stability purpose. The code is explicit in time using a \( 4^\text{th} \) order Runge-Kutta method. To capture stiff pressure waves induced by the plasma discharge, the hyper-viscosity technique developed by Cook and Cabot (2004) and Fiorina and Lele (2007) is employed.

The numerical configuration is shown in Fig. 2. The plasma discharge channel is assumed cylindrical with diameter \( D_y = 500 \) \( \mu \)m and height of \( L_d = 4 \) mm. Since \( D_y/L_d \ll 1 \), both the temperature and species axial gradients are neglected as in Naidis (2008), and a 2-D computational domain, perpendicular to the electrode tips, is considered as shown in Fig. 2. The computations are performed on a uniform mesh grid with a cell size of \( 10 \times 10 \) \( \mu \)m.

5 Results

5.1 – NRP discharges in air:

Figure 3-a shows the temporal evolution of the integrated values over the computational domain of total energy \( (E_{tot}) \), vibrational energy \( (E_{vib}) \) and energy deposited by the discharges \( (E_{discharge}) \), for a train of four NRP pulses. The results show that at each pulse \( (t = 0, 100, 200 \) and \( 300 \) \( \mu \)s) the energy deposited by the discharge goes either to translational or vibrational energy states. The vibrational energy is not relaxed into the total energy during the first pulse time interval. This means that, for low gas temperatures inside the discharge channel, the gas mixture properties are mostly influenced by the ultra-fast phenomena, whereas the vibrational energy is being accumulated over the discharges.

The vibrational energy relaxed into heating becomes more important with the increase of the gas temperature inside the discharge zone. As shown in Fig. 3-b, the maximum value of the gas temperature is higher at the beginning of each consecutive pulse. With the increase of the gas temperature, the gas density inside the discharge channel decreases. Therefore, for a constant discharge energy deposition per unit volume, the energy deposited per unit of mass increases with the increase of the gas temperature inside the discharge channel. The lower the gas density at the beginning of each discharge, the more efficient is the deposited energy on the increase of the gas temperature and on the increase of \( O \) mass fraction within the pulse characteristic time, as a result of the process described by Eqs. (7) and (13). The \( O \) atoms mole fraction, \( X_O \), increases along with the increase of the gas temperature, inside the discharge zone. This results in the decrease of \( \tau_{VT} \) given by Eq. (2), explaining the
temporal evolution of the source term $R^p_{TV}$ shown in Fig. 3-c and the consequent evolution of the vibrational energy, $E_{ vib }$, shown in Fig. 3-a.

Figure 3-d shows the temporal evolution of maximum gas pressure. During the ultra-fast period of the discharge, energy is deposited in a constant density gas. This leads to a fast increase of the gas pressure inside the discharge zone. The system then expands with the formation of a pressure wave that leaves the discharge zone and propagates with the speed of sound. The wave propagation velocity weakly depends on the gas temperature inside the discharge channel, depending mostly on the surrounding gas temperature (the pressure wave leaves the computational domain approximately at the same time instant, Fig. 3-d). This expansion phase also impacts the maximum gas temperature, early after each discharge, as shown in Fig. 3-b, by a sharp decrease of the maximum gas temperature. The results are in good agreement with the results presented in Xu et al. (2011) and Pancheshnyi et al. (2006).

5.2 – NRP discharges in methane-air mixture:

To study NRP discharges in reactive mixtures, 3 cases are compared. In all cases, the discharge frequency ($f = 10$ kHz) as well as the deposited energy ($E_{ discharge } = 0.8$ mJ per pulse), in each pulse, is kept constant. In case 1 the gas mixture is initially at rest whereas in case 2 and case 3 the gas flow is initialized with a homogeneous isotropic turbulent flow, characterized by a turbulent Reynolds number of 62 and 187, respectively.

5.2.1 – case 1 – Mixture ignition in quiescent flow condition:

Figure 4 shows the evolution of O atoms, OH and CH$_2$O radicals during the discharge time period of 50 ns. Along with the formation of O atoms, OH, CH$_2$O and other intermediate combustion species are formed. Both O and OH species are rapidly consumed after the first discharge and by the time the second discharge is applied, $t = 100$ µs, few O and OH species remain inside the discharge zone. In contrast, the CH$_2$O mass fraction increases over $\Delta t = 100$ µs. When the energy of the second discharge is deposited at $t = 100$ µs + 50 ns, the O and OH mass fractions drastically increase inside the discharge zone compared to the first discharge. This is explained by the increase of discharge energy per unit mass, as discussed in the previous section of NRP discharges in air.

The combined effect of the increase of combustion intermediate species and the increase of gas temperature inside the discharge zone, due to these first two NRP discharges, leads to mixture ignition. Figure 5 shows the 1-D longitudinal profiles of gas temperature during the discharges and during initial kernel propagation. During the transient regime of kernel propagation, the species mass fraction profiles evolve towards the laminar flame profiles as shown in Fig.6.
5.2.1 – case 2 and case 3 – Mixture ignition in turbulent flow in conditions:

Figure 7-a shows the temperature field at the beginning of the second discharge, \( t = 100 \, \mu s \), and at the end of this discharge, \( t = 100 \, \mu s + 50 \, ns \) for both case 2 and case 3. At the beginning of the discharge \( (t = 100 \, \mu s) \), the maximum mixture temperature inside the discharge zone is higher for case 2 than for case 3. This means that after the previous pulse, due to the higher turbulence intensity of case 3, the gas temperature inside the discharge channel is more convected and diffused around the discharge channel. As explained earlier for NRP discharges in air, the higher the gas temperature inside the discharge zone (and thus the smaller the gas density) the higher the impact of the deposited energy on the gas temperature increase. Therefore, in case 2, the gas temperature rises from \( T = 700 \) to \( 1500 \, K \), whereas in case 3, the gas temperature increase is less pronounced. In case 2, scalar fields inside the discharge zone are not much affected, yielding a homogeneous hot-spot formation during the discharge (Fig. 7-a, \( t = 100 \, \mu s + 50 \, ns \)), similar to conditions of case 1. However, in conditions of higher turbulent Reynolds number, scalar fields become highly non-homogeneous inside the discharge zone. In case 2, the mixture ignites right after the second NRP discharge, whereas in case 3, the mixture ignites only after three NRP discharges.

Figure 7-a and b show that, although turbulence does not have an effect during the discharge characteristic time \( (t_{per} = 50 \, ns) \), it modifies all the scalar fields in between discharges. As presented in Fig. 7-b, the vibrational energy is convected beyond the discharge zone where the mixture temperature and the oxygen concentration are low to induce a strong relaxation of the vibrational energy. The higher the turbulence intensity, the higher the convection and the diffusion of the vibrational energy beyond the zone where the plasma energy was deposited. This phenomenon also affects mass and radicals distribution inside the discharge zone. Depending on the pulse repetition frequency these turbulent fluctuations may lead to the failure of ignition.

Figure 8 shows the impact of turbulence on the temporal evolution of the maximum value of mixture temperature and of the integrated values of the mixture vibrational energy. It compares case 1 (quiescent flow), case 2 (turbulent flow, \( Re_\text{c} = 62 \)) and case 3 (turbulent flow, \( Re_\text{c} = 187 \)). For higher turbulence intensities the maximum value of the gas temperature drops faster between pulses. However, the low turbulence intensity of case 2 does not affect much the temperature inside the discharge zone. In this sense, case 1 and case 2 ignite with the same amount of deposited...
Figure 7 – a) mixture temperature field in 2 cases of initial turbulent flow conditions; b) vibrational energy field for the same cases. The graphic captures the moment when the second pulse is applied for both case 2 and case 3 and when the third pulse is applied for case 3.

Figure 8 – Temporal evolution of temperature and integral values of vibrational energy over the 2-D domain. The last graph shows the ratio between the vibrational energy that remains within an area with radius twice larger than the discharge radius and total vibrational energy in the domain. Initially all the vibrational energy remains inside this area for all cases, but as the times evolves, in case 3 turbulence convects this energy beyond the discharge zone and so this ratio decreases.

6 Conclusions

We have constructed a semi-empirical NRP discharge model that can be added to any multi-component reactive flow governing equations, in order to reduce the computational costs of NRP discharge-assisted combustion simulations while retaining the key physico-chemical mechanisms induced by nanosecond discharge pulses. A transport equation of vibrational energy is added to this system of governing equations. Along with this transport equation, four source terms are modeled based on experimental and numerical results. The model was analyzed for a train of NRP discharges in air and in a methane-air mixture for quiescent and turbulent flow conditions.

The results show that for low initial gas temperatures the energy deposited by the electric discharge
remains stored as gas vibrational energy. In these conditions, only the ultra-fast phenomena influence the thermodynamic and chemical properties of the gas mixture. As the gas temperature increases, the relaxation of the vibrational energy becomes more effective, leading to a faster increase of the gas temperature.

The results also show that in turbulent flow conditions, flow field characteristics should be taken into account when computing a train of NRP discharges in reactive mixtures. This may be critical in turbulent flows and reactive mixture conditions where the local values of species concentrations and temperature are governed by both hydrocarbon reactions and turbulence.

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