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The role of non-thermal electrons in flame acceleration

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Abstract

We examine in this work the effect of an external electric field on the propagation velocity of lean premixed flames, with the final goal of clarifying the relative importance of every of the three different mechanisms postulated in the literature to explain the effect of the electric field on flames: ionic wind, kinetic enhancement by non-thermal electrons and ohmic heating. The one-dimensional model proposed here expands the four-reactions scheme previously presented by [1] to include the effect of non-thermal electrons and activated neutral molecules on flame acceleration. Two additional reactions are included in the model to complete a minimum set of six elementary reaction capable of qualitatively reproduce the results observed in classical [2] and recent [3, 4] experiments. The limit of weakly ionized plasmas is used to integrate the Boltzmann equation and to derive an explicit expression for the electron temperature proportional to the square of the electric field. The numerical integration of the conservation equations gives the flame propagation velocity for a given set of parameters. The results reveal the importance of the electric field polarity on flame acceleration, finding faster flames for positive electric fields than for equally intense negative fields. At low-intensity fields, our results indicate that the ionic wind, and the associated redistribution of the charged particles, is the main mechanism inducing flame acceleration. In more intense fields, the combined effect of the ionic wind and the heat transfer from the high-temperature electrons to the background gas induces a significant increase in the temperature field upstream and downstream of the flame front. Associated with this temperature increase, relevant changes on the flame speed are computed for positive, intense electric fields, while only modest flame accelerations are observed for equally intense negative fields, behavior that reproduces qualitatively the measurements by [4]. The reduced sensitivity to an external electric field when the mixture approaches stoichiometry, observed experimentally by [2] and [5], is also reproduced by the model proposed in this work.

Keywords: Electric field, Flame speed, Ohmic heating, Ionic wind, Activated species

1. Introduction

Although the effect of electric fields on flames has been known since since 1814 [6], there is still a great controversy to explain the disagreement of both magnitude and type of response between the different experimental results published in the literature. As an example of this debate, consider experiments on flame speed measurements under the effect of an electric field. Among the experimental data published, we find slight flame deceleration [7, 8], slight flame acceleration [9, 10, 11, 12] and strong flame acceleration of up to 200 % [2, 4, 13].

Three different mechanisms have been postulated to explain the effect of the electric field on flames: ionic wind, kinetic enhancement by non-thermal electrons and ohmic heating. Chemical effects via activation of reactions has been identified by [2, 13, 9] as the main mechanism for flame acceleration in planar flames while body forces (ionic wind) are, apparently, solely responsible for the observed response in diffusion and premixed flames in complex geometries [14, 15, 5, 16]. The effect on the flame speed of the heating of the background gas as a consequence of the higher temperature of the non-thermal electrons (ohmic heating) has been reported as important, among others, in the microwave induced flame speed enhancement experiments carried out by [17, 18] and in the computations by Bradley & Ibrahim [19].

In order to identify the relative importance of each of these three mechanisms, we consider in this paper a one-dimensional model of a lean premixed flame under the influence of a longitudinal electric field. The electric field is oriented in the direction of the gas flow and can have positive or negative sign, indicating different electric field polarities. The model is defined by the set of conservation equations and chemical reactions which should reflect the behavior of a lean premixed flame subjected to an electric field. To model the interaction between the flame and the

i _{SD}	Reaction	Rate
I	$F+Z \Rightarrow 2 Z$	$\Omega_I = A_I \frac{\rho^2}{W_F W_Z} Y'_Z Y'_F \exp\left(-\frac{E_I}{RT}\right)$
II	$Z+M \Rightarrow P + M + q_{II}$	$\Omega_{II} = A_{II} \frac{\rho^2}{W_Z \bar{W}} Y'_Z$
III	$Z+M \Rightarrow Z^+ + e^- + M$	$\Omega_{III} = A_{III} \frac{\rho^2}{W_Z \bar{W}} Y'_Z \exp\left(-\frac{E_{III}}{RT}\right)$
IV	$Z^+ + e^- \Rightarrow P + q_{IV}$	$\Omega_{IV} = A_{IV} \frac{\rho^2}{W_{Z^+} W_{e^-}} Y'_{Z^+} Y'_e$
V	$Z+M^* \Rightarrow P + M + q_{II}$	$\Omega_V = \Lambda' A_{II} \frac{\rho^2}{W_Z \bar{W}} Y'_Z Y_{M^*}$
VI	$e^- + M \Rightarrow M^* + e^-$	$\Omega_{VI} = C \nu_{eh} n_e \exp\left(-\frac{E_{VI}}{RT_e}\right)$

Table 1: List of reactions

electric field, we need to include in the chemistry model a radical that can be ionized at high temperatures to give ions and electrons that can be affected by the electric field. Additionally, the model should include a mechanism to describe the transfer of energy from the external electric field to the fluid and to account for the energy absorbed by the neutral molecules that become vibrationally or rotationally excited as a consequence of the collisions with high-energy electrons.

Because of its simplicity and ability to mimic the behavior of real hydrocarbons [20], we chose the two-step, chain-branching chemistry model developed originally by [21, 22] as the starting point. Two more reactions need to be incorporated to account for the ionization of the intermediate radical (the chemi-ionization of the intermediate radical, reaction III) and its subsequent recombination with the free electrons (dissociative recombination, reaction IV) generated in the high temperature region of the flame. By including these two additional steps, Sánchez-Sanz et al. [1] studied the effect of the ionic wind on premixed flame acceleration, finding flames that were around 15% faster when an external electric field was applied. A similar mechanism has been used recently by Patyal, Kyritsis and Matalon [23] to study the effect of an electric field on the burning characteristics of a spherically symmetric fuel drop.

The presence of an electric field modifies the electron energy distribution function and elevates the temperature of the electrons, as dictated below by the equation (4). High energy electrons and ions might produce active radicals and electronically and vibrationally excited molecules (e.g., $N_2(v)$). According to [24], reaction

is the most likely way in which a field can affect reactions. Subsequently, a vibrationally excited nitrogen molecule can transfer vibrational quanta to other molecules (in particular, O_2)

Such vibrational excitation of oxygen molecules can accelerate chemical reactions, in particular, radical consumption, and, accordingly, the combustion process as a whole. In our model, reactions (1) and (2) are modeled by reaction VI. Reaction V accounts for the accelerated consumption of radicals induced by activated species by adding a multiplying factor $\Lambda > 1$ to the frequency factor of reaction II. The concentration of activated species is moderated by the energy transferred from the electrons to the neutral species when the two particles collide. Only very energetic electrons can transfer sufficient energy to excite heavy molecules. For example, only electrons with energy above 0.289 eV (electron temperature above 3353 K) or 11.548 eV (electron temperature above 134,000 K) can excite the first vibrational mode of nitrogen molecules [25] or raise argon to its first electronic state [26], respectively.

The ohmic heating of gases is caused by electrons losing energy to neutral molecules with the corresponding increase in temperature and reactivity of the mixture. This energy transfer is proportional to the rate of collisions between the electrons and neutral gas molecules and will be taken into account by including the corresponding term into the energy equation.

2. Formulation

The reaction zone of a hydrocarbon flame exhibits an unusually high degree of ionization, due to chemiionization reactions. The degree of ionization usually depends on the fuel burned but it is generally accepted that the flame constitutes a weakly ionized gas in which the concentration of electrons (around 10^{16} particles/m³ [27]) is much smaller than the concentration of neutral particles [28]. Most of the previous modeling efforts considered thermal electrons (electron temperature equal to the background gas temperature) [1, 15, 29], a hypothesis that facilitates the numerical integration of the problem but limits the applicability of the results. To cover this gap, we consider, in this work, the effect of non-thermal electrons on the flame acceleration of a premixed flame.

At low electric fields, E' , the energy of electrons is not enough to excite the mixture components and the ionic wind can be used, almost exclusively, to explain the interaction between the flame and the external field [24]. As the intensity of the field E' increases, the gas excitation by electron-impact becomes important. This process leads to the creation of chemically active species and new interaction mechanism that should be taken into account. In lean flames, the most probable collision is between the electrons and molecules of the neutral gas, the most abundant component of the gas mixture. As a consequence of this, the collision frequency between the electron and all the heavy particles can be written, in the first approximation, as $\zeta_{eH} = \sum_h \zeta_{eh} \simeq \zeta_{eM}$ being ζ_{eM} the electron-neutral gas particles collision frequency.

After an effective collision, the electron transfers energy to the neutral gas M creating an activated molecule M^* . This activated species, that will be treated as a distinct species, may react with the intermediate radical Z to give products and release heat, as indicated by reaction V.

Strictly speaking, these collision events will remove energy from the electron population, lowering their temperature. However, we shall assume that the high activation energy of this process makes these collisions immensely less common than the ordinary inelastic scattering between electrons and heavy particles. Therefore, in pursuit of succinct formulation, the effect of the energy lost by the electrons when activating the neutral gas will be omitted in the calculation of the electron temperature T'_e .

The calculation T'_e implies the integration of the Boltzmann equation to obtain the electron energy distribution function f^0 . As shown by [30], in a weakly ionized plasma the electron-electron collision frequency ζ_{ee} is much smaller than the electron-heavy particles collision frequency ζ_{eH} , such that $\zeta_{ee} \ll \sum_h \zeta_{eh}$. In this limit, the Boltzmann equation can be integrated to give the electron energy distribution function

$$f^0(C) = f^0(0) \exp \left\{ - \int_0^C \frac{m_e - C' dC'}{k_B T + \frac{e^2}{3m_e^2 - \zeta_{eH}} \frac{E^2}{\sum_h \zeta_{eh}/m_h}} \right\} \quad (3)$$

where k_B is the Boltzmann constant and C is the velocity of the electrons. The electron-heavy particles collision frequency $\zeta_{eH} \simeq \zeta_{eM}$ and $\sum_h \zeta_{eh}/m_h \simeq \zeta_{eM}/m_M$ are assumed constant. The integration constant

$$f^0(0) = \left\{ (m_e/2\pi) / \left[kT + e^2/(3m_e^2 - \zeta_{eH}) E^2 / \sum_h (\zeta_{eh}/m_h) \right] \right\}^{3/2}$$

is obtained from the normalization condition $\int_0^\infty f^0(C) C^2 dC = 1/4\pi$. Once the electron energy distribution function is known, the calculation of the electron temperature $T'_e = 4\pi m_e / (3k_B) \int_0^\infty C^4 f^0(C) dC$, yields

$$T'_e = T' + \frac{1}{3} \frac{e^2 m_M}{k_B m_e^2 \zeta_{eH}} E'^2 \quad (4)$$

Notice that the temperature difference between the electrons and the background gas $T'_e - T'$ is proportional to the square of the electric field, as predicted by Fridman [31] in an isobaric environment such as the one considered here. Only in the case of small values of E' the temperature of electrons and heavy particles approach to each other. The accuracy of the Lorentzian limit for weakly ionized plasmas has been tested recently by [32] in the calculation of the transport properties in multicomponent plasmas.

The kinetics of combustion can be affected by an external electric field by changing the reaction rates of some of the reactions that involve activated species. In our case, this effect is modeled in reaction V by introducing a factor Λ , what

makes this reaction significantly faster than reaction II , where the radical recombines to release heat with the help of a non-activated third body. According to [33], this factor could be as large as 10^3 .

Once the electron temperature is calculated, in the constant density approximation considered here, the problem reduces to the integration of energy and mass transport equations for neutral and charged species. Unlike neutral gases, where diffusion is controlled by Fick's law, the presence of an electric field can change the way in which the particles are redistributed in an inhomogeneous mixture. Even when no external electric field is applied, a displacement of charged particles would create a charge imbalance that, in turn, would induce an electric field opposing the charge displacement. This effect has been taken into account in the calculation of diffusion velocities, as it is specified below in equations (12) and (13).

The diffusion coefficient of the neutral species is considered constant, being D_F and D_Z the diffusion coefficients of fuel and radical Z , respectively. On the other hand, Belhi et al [15] introduced, based on the work by Delcroix & Bers [34], the following expression for the ratio between the diffusion coefficient of electrons and ions $D_{e-} = D_{Z+}(m_{Z+}/m_{e-})^{1/2}$ where m_{Z+} and m_{e-} are the mass of a single positive ion and electron respectively.

The mobility of a charged particle ν_i is defined as the ratio between its drift velocity and the electric field strength. The strict calculation of the mobility of ions ν_{Z+} and electrons ν_{e-} would imply the appropriate assessment of the effect of the temperature and concentration changes on ν_i [35]. Nevertheless, and for the sake of simplicity, we will assume hereafter constant cation and electron mobilities. Furthermore, we can write the ratio between the mobilities of electrons and ions by using the Einstein relationship $\nu_{e-} = \nu_{Z+}(m_{Z+}/m_{e-})^{1/2}$ given in [15].

As a summary of the exposed above, we introduce in the energy and mass conservation equations the non-dimensional temperature $T = (T' - T_0)/(T_c - T_0)$ and spatial coordinate $x = x'/(D_T/S_L)$ and the scaled mass fractions of fuel $Y_F = Y'_F/Y_{F0}$, radical $Y_Z = Y'_Z/[Y_{F0}(W_Z/W_F)]$, positive ions $Y_{Z+} = Y'_{Z+}/[Y_{F0}(W_Z/W_F)]$, electrons $Y_{e-} = Y'_{e-}/[Y_{F0}(m_{e-}/m_{Z+})(W_Z/W_F)]$ and activated neutrals $Y_{M*} = Y'_{M*}/(Y_{F0}\bar{W}/W_F)$ to yield the non-dimensional conservation equations

$$\frac{dT}{dx} = \frac{d^2T}{dx^2} + R_\Omega \mu Y_e \tilde{\zeta}_{eh} \tilde{Y}_{F0} \frac{m_{e-}}{m_{Z+}} (T_e - T) + \mu Q \left(\omega_{II} + \omega_{IV} \frac{q_{IV}}{q_{II}} + \omega_V \right) \quad (5)$$

$$\frac{dY_e}{dx} = \mu (\omega_{III} - \omega_{IV}) - \frac{d}{dx} (Y_e u_e) \quad (6)$$

$$\frac{dY_F}{dx} = \frac{1}{Le_F} \frac{d^2Y_F}{dx^2} - \mu \omega_I \quad (7)$$

$$\frac{dY_{Z+}}{dx} = \mu (\omega_{III} - \omega_{IV}) - \frac{d}{dx} (Y_{Z+} u_{Z+}) \quad (8)$$

$$\frac{dY_Z}{dx} = \frac{1}{Le_Z} \frac{d^2Y_Z}{dx^2} + \mu (\omega_I - \omega_{II} - \omega_{III} - \omega_V) \quad (9)$$

$$\frac{dY_{M*}}{dx} = \frac{1}{Le_{M*}} \frac{d^2Y_{M*}}{dx^2} + \mu (\omega_{VI} - \omega_V), \quad (10)$$

to be integrated with boundary conditions $T = Y_i = 0$ for $i \neq F$ and $Y_F = 1$ at $x \rightarrow -\infty$ and $dT/dx = dY_i/dx = 0$ at $x \rightarrow \infty$. The solution of the problem provides the eigenvalue

$$\mu = \frac{\rho A_{II} D_T}{S_L^2 \bar{W}} \quad (11)$$

which determines completely the flame propagation velocity S_L . In the previous system of equations $Le_F = Le_Z = Le_{M*} = 1$ are the Lewis numbers of the fuel, the radical and the activated neutral species, $u_e = u'_e/S_L$ and $u_{Z+} = u'_{Z+}/S_L$ are the displacement velocity of electron and positive ions.

According to Mitchner & Krugger [30] the displacement velocity of electrons $u_e = u'_e/S_L$ and ions $u_{Z+} = u'_{Z+}/S_L$ can be calculated as

$$Y_e u_e = -\mu^{1/2} \frac{\nu_e}{\nu_{Z+}} Y_e (E_i + R_I E_0) - \frac{1}{Le_Z} \frac{D_{e-}}{D_{Z+}} \frac{d}{dx} \left(Y_e \left(T_e + \frac{1-\gamma}{\gamma} \right) \right) \quad (12)$$

$$Y_{Z+} u_{Z+} = \mu^{1/2} Y_{Z+} (E_i + R_I E_0) - \frac{1}{Le_Z} \frac{d}{dx} \left(Y_{Z+} \left(T + \frac{1-\gamma}{\gamma} \right) \right) \quad (13)$$

where E_i and E_0 are the auto-induced and the externally-applied electric field, respectively, defined in non-dimensional form as $E_j = \nu_{Z+} E'_j / (\rho A_{II} D_T / \bar{W})^{1/2}$. The auto-induced electric field E_i is calculated by integrating

$$\frac{dE_i}{dx} = \tilde{Y}_{F_0} \frac{\mu^{1/2}}{\varepsilon} (Y_{Z+} - Y_{e-}) \quad E_i(x \rightarrow -\infty) = 0 \quad (14)$$

where the non-dimensional permittivity is $\varepsilon = (\varepsilon_0 A_{II} / e \nu_{Z+}) / [W_Z / W_F]$. In the limit $\varepsilon \rightarrow \infty$, E_i is small and can be neglected, in the first approximation.

In equations (5), (12) and (13) we have included two artificial coefficients $0 \leq R_\Omega \leq 1$ and $0 \leq R_I \leq 1$ to numerically study the effect of the ohmic heating and of the ionic wind in flame acceleration. As will be show below, the presence or not of these terms in the equations modify significantly the effect of the electric field on the flame and will be used to study how important they are in explaining the experimental observations.

As we showed above in (4) for a weakly ionized plasma, the temperature of the electrons under the effect of an externally applied electric field E_0 can be calculated as

$$T_e = T + \frac{2}{3Le_{Z+}} \left(\frac{m_{Z+}}{m_{e-}} \right)^{3/2} \frac{1}{\tilde{\zeta}_{eH}} (E_i + E_0)^2, \quad (15)$$

with $T_e = (T'_e - T_0) / (T_c - T_0)$, the non-dimensional electron temperature, and $\tilde{\zeta}_{eH} = 2\zeta_{eH} W_Z W_{Z+} / (\rho W_F A_{II})$, the non-dimensional collision frequency. The branching temperature T_c , used to define the non-dimensional temperatures, is the minimum temperature of the gas required for the chain branching reaction to take place [1, 20], and can be calculated by imposing $\Omega_I = \beta^2 \Omega_{II}$. Also, the following non-dimensional parameters appear in the above formulation: the Zel'dovich number $\beta = E_I (T_c - T_0) / (R T_c^2) = 10$, with E_I the activation energy of reaction I and R the ideal gas constant, and the heat release parameter $\gamma = (T_c - T_0) / T_c = 0.7$. The dimensionless heat of reaction $Q = q_{II} Y_{F_0} / [c_p (T_c - T_0) W_F]$, with q_{II} representing the total heat released from reactions II, is a parameter of the problem and its value will be varied to study its effect on flame acceleration. Notice that changes of the parameter Q can be understood as slight modifications on the fuel mass fraction of the mixture Y_{F_0} , so that larger values of Q would imply richer mixtures.

The non-dimensional reaction rates are written as

$$\omega_I = \beta^2 Y_Z Y_F \exp \left\{ \beta \frac{T - 1}{1 + \gamma(T - 1)} \right\} \quad (16)$$

$$\omega_{II} = Y_Z \quad (17)$$

$$\omega_{III} = \beta^2 \frac{\mathcal{A}}{\tilde{Y}_{F_0}} Y_Z \exp \left\{ (\beta + \Delta\beta_{III}) \frac{T - 1}{1 + \gamma(T - 1)} \right\} \quad (18)$$

$$\omega_{IV} = \mathcal{B} \tilde{Y}_{F_0} Y_{Z+} Y_{e-} \quad (19)$$

$$\omega_V = \Lambda \tilde{Y}_{F_0} Y_Z Y_{M^*} \quad (20)$$

$$\omega_{VI} = \beta^2 \mathcal{F} \tilde{\zeta}_{eH} Y_e \exp \left\{ (\beta + \Delta\beta_{VI}) \frac{T_e - 1}{1 + \gamma(T_e - 1)} \right\} \quad (21)$$

with $\mathcal{A}, \mathcal{B}, \mathcal{F}, \Delta\beta_j$ and Λ defined in table 2. In real flames, the heat released through the termination reaction q_{II} is different to that released through the dissociative recombination step $q_{IV} \neq q_{II}$. Nevertheless, and for the sake of simplicity, we will assume hereafter $q_{IV} = q_{II}$.

In table 2 we summarized the definition of the parameters introduced in the formulation and the values used in the computations shown below.

3. Numerical method

The problem defined by Eqs. (5)- (10), with the corresponding boundary conditions, was solved numerically to compute the eigenvalue μ and the profiles of temperature and species in a uniform grid spanning from $x_{min} = -100$ to $x_{max} = 100$. The spatial derivatives were discretized using second order, three- point central differences in a grid formed by $N = 2500$ points, what gives a minimum spacing of $dx \simeq 0.01$ at $x = 0$. A 20 % increase in the number of

Parameter	Definition	Values
Pre-exponential factor	$\mathcal{F} = e^{-\Delta\beta_{VI}/\gamma} \frac{C/2}{m_{Z^+}} \frac{A_{II}}{A_I} \frac{W_F}{W_{Z^+}}$	$\mathcal{F} = 2.5 \times 10^{-4}$
Chemistry enhance factor	$\Lambda = \Lambda' \frac{W_Z}{W_F}$	$\Lambda = 80$
Collision frequency	$\tilde{\zeta}_{eh} = 2\zeta_{eh} \frac{W_z W_{z^+}}{W_F \bar{W}} \frac{\bar{W}}{\rho A_{II}}$	$\tilde{\zeta}_{eh} = 2560$
Electric field	$E = \nu_{Z^+} \frac{E'}{(\rho A_{II} D_T / \bar{W})^{1/2}}$	$-2 < E < 2$
Heat of reaction	$Q = \frac{q_{II} Y_{F_0}}{c_p (T_c - T_0) W_F}$	$Q = 5.0$
Fuel mass fraction	$\tilde{Y}_{F_0} = Y_{F_0} \frac{m_M}{m_{Z^+}}$	$\tilde{Y}_{F_0} = 0.25$
Zel'dovich number	$\beta = E_I (T_c - T_0) / (R T_c^2)$	$\beta = 10$
Lewis number	$Le_i = D_i / D_T$	$Le_i = 1$
Mass ratio	m_{Z^+} / m_{e^-}	$\frac{m_{Z^+}}{m_{e^-}} = 64$
Heat release parameter	$\gamma = (T_c - T_0) / T_c$	$\gamma = 0.7$
Frequency factor chemiionization	$\mathcal{A} = \frac{A_{III}}{A_I} \frac{W_F}{W_Z} e^{-\Delta\beta/\gamma}$	$\mathcal{A} = 2.5 \times 10^{-5}$
Frequency factor recombination	$\mathcal{B} = \frac{A_{IV}}{A_{II}} \frac{W_Z}{W_F}$	$\mathcal{B} = 40$
Differential Activation Energy	$\Delta\beta_j = \gamma \frac{E_j - E_I}{R_g T_c}$	$\Delta\beta_{III} = 1$ $\Delta\beta_{VI} = 1$
Permittivity	$\epsilon = \frac{\epsilon_0 A_{II}}{e \nu^+} \frac{W_F}{W_Z}$	$\epsilon = 2.5$

Table 2: Characteristic values of the parameters used in the figures shown below.

points was used in some cases to test the grid independence of the numerical solution.

The eigenvalue μ and the profiles of temperature and species were computed using an iterative method based on a GaussSeidel procedure with over-relaxation that takes advantage of the invariance of the equations to a translation in the coordinate x . Using this property, a random value of temperature T^* is forced at the grid point x^* such that $T(x^*) = T^*$, what gives an additional condition that allows the calculation of the eigenvalue μ^k at the iteration k from Eq. (5). To avoid the divergence of the method, we used a relaxation parameter α so that the value of the eigenvalue used at the next iteration $k + 1$ is given by $\mu^{k+1} = \alpha\mu^k + (1 - \alpha)\mu^{k-1}$. Typical values of above mentioned parameters are $\alpha = 0.5$ and $T^* = 0.65$. A comprehensive description of the numerical procedure outlined above can be found in [36].

4. Results

4.1. Heat release Q

Figure 1: **a)** Variation of the normalized eigenvalue $(\mu_0/\mu)^{1/2}$ with the heat release parameter for $\Lambda = 80$ and $Q = 3$ (solid curves) and $Q = 5$ (dot-dashed curves). The symbols indicate different values of the kinetic enhancement parameter: $\Lambda = 4$ ($\square$) and $\Lambda = 200$ ($\circ$). The reduced sensitivity of the normalized eigenvalue with an increasing Q is shown in the inset for $E = 0.5$. **b)** Spatial distribution of the ohmic heating term $\mu \bar{\zeta}_{eh} \bar{Y}_{F0} \frac{m_e}{m_{Z^+}} Y_e (T_e - T) / Q$ for $Q = 3$ (solid line) and $Q = 5$ (dot-dashed lines). **c)** Spatial distribution of the reactive terms $\mu(\omega_{II} + \omega_{IV} + \omega_V)$ and the scaled temperature T/Q for $Q = 3$ (solid line) and $Q = 5$ (dot-dashed lines).

The system of equations described above in (5)-(10) can be integrated once to give the temperature downstream of the flame T_∞ , yielding

$$\frac{T_\infty}{Q} = 1 + \mu \frac{\bar{\zeta}_{eH}}{Q} \int_{-\infty}^{\infty} Y_e (T_e - T) dx. \quad (22)$$

when no fuel leakage is found and $q_{IV} = q_{II}$. The first term in the right hand side of this equation represent the temperature increase caused by the heat released during the recombination of the radicals Z and Z^+ . The second term accounts for the temperature rise associated to the ohmic heating. In the presence of none or weak electric fields, the heat release parameter Q is the maximum temperature achieved by the flame when all the fuel is consumed. Near the flammability limit $Q \simeq 1$, when appreciable fuel leakage is found, colder flames $T_\infty < Q$ are expected [36]. In flames under the influence of strong electric fields, the ohmic heating heats up the gas above the maximum temperature Q achieved by chemical recombination exclusively. As become evident from equation (22), the contribution of the

last term is proportional to the concentration of electrons and to the collision frequency, anticipating higher flame temperatures when the production of electrons via chemiionization is promoted.

With the rest of the parameters fixed, we analyze the influence of the heat release parameter Q in figure 1 by plotting the evolution the eigenvalue normalized by the value of the eigenvalue with no electric field $(\mu_0/\mu)^{1/2} = S_L/S_{L_0}$ for different values of the external electric field E_0 . The computations show the importance of the electric field polarity, indicating stronger flame sensitivity for $E_0 > 0$ as the more abundant concentration of high temperature electrons in the low temperature region of the flame heats up the gas appreciably above the background temperature. With $E_0 < 0$, the concentration of electrons in this regions is reduced and the effect on the temperature is less remarkable, as can be seen in Fig. 1. Even when the overall effect on the temperature is subtle, it is sufficient to fourfold, for $Q = 3$, or double, for $Q = 5$ the flame speed for sufficiently large electric fields. A similar behavior has been previously reported in the experiments by Murphy et al [4], Karnadi and Dunn-Rankin [37], Volkov et al. [3] and Jagers and Von Engel [38].

Unlike the previous, transport-based models (see, for example, [1]), the new model exhibits reduced sensitivity to the an external electric field for higher values of Q as the heat introduced by electron thermalization, proportional to $Y_e(T_e - T)/Q$, becomes comparatively less important for more energetic mixtures, as shown in Fig. 1b) for $E_0 = 0.5$. As a consequence of this, the heat added ahead of the flame, associated mainly to the ohmic heating reduces. The formation of Z and Z^+ through the temperature-dependent reactions I and III is then limited what, in turns, restricts the heat released through reactions II , IV and V (see Fig. 1c)). This result is also consistent with the results reported by Jagers and Von Engel[2] and by Murphy et al. [4] for pre-mixed methane air-flames over a range of equivalence ratios.

Vibrationally-excited molecules can accelerate chemical reactions by enhancing radical consumption. In our model, reaction V accounts for the accelerated consumption of radicals induced by activated species by means of the multiplying factor Λ in reaction II. The larger the value of Λ , the faster the radical Z will be consumed. Figures 1 and 2 summarize the effect of Λ on the propagation velocity and on the structure of the solution. As shown in Fig. 1, the flame becomes slower as Λ is increased, so much so when Q is large. As the parameter Λ increases, as shown in Fig. 2, the heat is released more rapidly and the temperature jump takes place more abruptly, limiting the upstream diffusion of energy and the temperature rise in the low temperature region ahead of the flame.

Although the evident changes on the concentrations of the species and on the temperature observed in Fig. 2 the effect of Λ is small when negative electric fields are applied $E_0 < 0$, with flame propagation velocities almost identical to the case in which no electric fields are applied. On the contrary, positive electric fields induce the diffusion of electrons upstream of the flame, heating up the mixture ahead of the reaction region and accelerating the flame. This result is in conflict with the results obtained by [2, 9, 13], who considered chemical effects via activation of reactions as the most important mechanism to explain flame speed enhancement. Possibly, the differences between the computations and the experiments are associated with the way the external field is applied in the experiments modifying, consequently, the interaction between the field and the chemistry. However, clearly more research is needed to clarify the way in which the external field modify chemical reactions.

4.2. The effect of the ionic wind R_I

The term ionic wind is used here to describe the driving force that favors the diffusion of electrons and positive ions towards a preferential direction determined by the sign of the electric field E_0 . This effect is quantified through the first term of the right hand side of equations (12) and (13) for electrons and positive ions, respectively. In the simple model considered here, we do not consider the transfer of momentum from the electrons to the neutral particles.

To study the influence of the ionic wind on the changes of the flame speed reported both numerically and experimentally, we artificially include the parameter R_I in equations (12) and (13). Therefore, when $R_I = 0$, the field-induced diffusion is not considered and the ohmic heating is to be blamed in the around 40 % flame speed enhancement shown in Fig. 3. A similar change on flame speed was obtained experimentally by [17] after eliminating the effect of the ionic wind by exciting a premixed methane flame with 50 Mhz microwaves pulses.

Even when the ionic wind is not considered, the polarity of the external electric field E_0 modifies the magnitude of the total electric field $E = E_i + E_0$, as shown in the inset of figure 3 for $E_0 = (0.1, 0, -0.1)$, what results in the slight anisotropy observed between negative and positive electric fields in the $(\mu_0/\mu)^{1/2}$ versus E_0 curve shown in Fig. 3.

As R_I approaches unity, the influence of field-induced preferential diffusion becomes more evident, leading to a remarkable flame speed enhancement for positive, moderately large fields.

Profiles of temperature, electrons and positive ions mass fractions and heat released obtained by integrating the above

Figure 2: **a)** Electrons and positive charged species Z^+ mass fraction and **b)** temperature T/Q and reactive terms $\mu(\omega_{II} + \omega_{IV} + \omega_V)$ for $\Lambda = 4$ (dashed curves), $\Lambda = 80$ (solid curves) and $\Lambda = 800$ (dot-dashed curves). In the left plots $E_0 = -1$ while in the right plots $E_0 = 1$.

system of equations with $R_I = 0$ are represented in Fig. 4 for $E_0 = -0.5$ and $E_0 = 0.5$ and in Fig. 5 for $E_0 = -1.5$ and $E_0 = 1.5$. In both figures, the profiles are compared with those obtained with $R_I = 1$.

For small fields intensities, the absence of the ionic wind $R_I = 0$ induces substantial changes in the local concentration of charged particles but only subtle differences in the temperature profiles are found downstream of the reaction region as a consequence of the ohmic heating. The flame acceleration for $|E_0| = 0.5$ is still modest. More intense fields $|E_0| = 1.5$ modify the temperature upstream and downstream of the reaction region, changing the rate of formation of the radical Z and Z^+ through the temperature-dependent reactions I and III. For $E_0 = -1.5$, the absence of the ionic wind increases the temperature and makes the flame faster than its counterpart with $R_I = 1$. The trend is reversed for $E_0 = 1.5$, when the ionic wind emerges as an important factor to allocate the high-temperature electrons in the reaction region, favoring the formation of Z and Z^+ .

4.3. The effect of the ohmic Heating R_Ω

Charged particles generated in the high temperature region of the flame are accelerated by the electric field. When they collide with the neutral particles, an amount of energy proportional to the square of the electric field E is released, as indicates the second term in the right hand side of equation (5). To quantify the importance of this term on flame acceleration, we have calculated the flame speed modifying the value of the parameter R_Ω added artificially in equation (5), such that when $R_\Omega = 1$ we take full account of this term and when $R_\Omega = 0$ the ohmic heating is neglected. We summarize the results of our computations in figure 3 where we plot the normalized flame speed for several values of R_Ω . When $R_\Omega = 0$, we recover the results reported by [1] when non-thermal electrons were not considered.

The changes in the flame speed reported in figure 3b are associated with the temperature of the electrons. After been created in the high temperature region through reaction III, the electrons are pushed by the electric field towards the low temperature region located upstream of the flame. When the ohmic heating is not considered $R_\Omega = 0$, the electrons do not transfer energy to the background gas and the temperature shift does not occur, as shown in figures 4 and 5. The effect of the ohmic heating is important for positive and negative electric fields, but its role becomes paramount for $E_0 > 0$. According to figures 3, when non-thermal electrons are not considered, the acceleration of the flame reaches a maximum at $E_0 = E_{max} \simeq 1.5$ for $Q = 5$, behavior not observed in the experiments. After thermal electrons are included, the speed of the flame increases continuously with the intensity of the electric field, as predicted by the experiments [4].

The combined effect of the ionic wind and the ohmic heating modifies both the temperature and the minor species profiles. When mild electric fields are applied, the change in the profiles Fig. 4 are subtle, but they become more evident for $E_0 = 1.5$ when the temperature is increased both upstream and downstream of the reaction region as a consequence of the combined effect of the ionic wind and the ohmic heating.

When considered separately, ionic wind and ohmic heating can only accelerate the flame around a 40 %. This result is in agreement with the experiments by [17, 18], where the effect of the ohmic heating is isolated by using microwave pulses to excite the flame. The fast 50 MHz alternating field does not allow the electrons to migrate away from the reaction region where they were created, limiting the influence of the field to the region where the electrons cluster in larger concentrations. This extra energy absorbed by the flame is then conducted upstream the reaction front, accelerating the flame propagation and, eventually, increasing the flammability limits [18]. According to our results, this effect is promoted when a positive DC electric field is used to influence the flame as a consequence of the combined effect of the ionic wind and of the ohmic heating. Additionally to the conduction of heat, the actual diffusion of electrons to the region immediately upstream of the reaction region elevates the temperature of the background gas, enhancing the formation of radicals and charged species through the temperature-dependent reactions I and III.

Figure 3: **a)** Variation of the normalized flame speed with the electric field for several values of the ionic wind parameter R_I keeping $R_\Omega = 1$. In the inset, we compare the spatial variation of the electric field when the ionic wind terms are switched off $R_I = 0$ (black curves) and when $R_I = 1$ (gray curves) for $E_0 = (-0.1, 0, 0.1)$. **b)** As in a) but changing the ohmic heating parameter R_Ω but keeping $R_I = 1$.

5. Conclusion

The effect of an external electric field on the propagation velocity of a laminar premixed flame has been examined. The one-dimensional model proposed here, based on the detailed combustion mechanisms used by [33, 27, 39], includes the ionization of the intermediate radical and its subsequent recombination with free electrons and considers the production of active radicals. Unlike previous modeling efforts, the model includes non-thermal electrons with the electron temperature calculated using an explicit expression derived from the integration of the Boltzmann equation in the limit of weakly ionized plasmas.

From the calculations carried out in this article, we have isolated the different interaction mechanisms between the electric field and the flame. To do so, we have artificially switched off in the equations the terms controlling the ohmic

Figure 4: **a)** Electrons and positive charged species Z^+ mass fraction and **b)** temperature T/Q and reactive terms $\mu(\omega_{II} + \omega_{IV} + \omega_V)$ for $R_I = 0$ (dashed curves), $R_\Omega = 0$ (dot-dashed lines) and $R_I = R_\Omega = 1$ (solid lines). In the left plots $E_0 = -0.5$ while in the right plots $E_0 = 0.5$.

Figure 5: Same as in Fig. 4 but with $E_0 = -1.5$ in the left plots and $E_0 = 1.5$ in the right plots.

heating and driving forces. According to our calculations, the ionic wind is responsible of flame acceleration for low-intensity electric fields, when the redistribution of electrons and positively charge particles enhance flame propagation. For more intense electric fields, the heat transfer from the electrons to the background particle favors the formation of radicals, what, in turn, speeds up flame propagation. The interaction between the two mechanisms for positive, intense fields can double the flame speed.

The accelerated consumption of radicals induced by activated species is taken into account by including a factor Λ on the frequency factor of the reaction controlling the consumption of the intermediate radical. The larger is this parameter, the more influence the activated species have in the combustion chemistry. According to our calculations, when Λ is increased, slower flame speed are computed, more so as the intensity of the electric field is increased. Our conclusions is in conflict with previous studies [2, 9, 13] that identified the chemical effects via activation of reactions as one of the main mechanisms for flame acceleration in planar flames. Possibly, the differences we found are associated with the way the external field is applied in the experiments modifying, consequently, how the external field interacts with the chemistry.

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