



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 a laminar, one-dimensional and lean premixed flame, with the final goal of clarifying the relative importance of each of the three different mechanisms postulated in the literature to explain the effect of electric fields 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 Sánchez-Sanz, et al. (2015) 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 (Jaggers, and Von Engel, (1971).) and recent (Volkov et al., 2013; Murphy, et al., 2014.) 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 Murphy et al. (2014). The reduced sensitivity to an external electric field when the mixture approaches stoichiometry, observed experimentally by Jaggers, and Von Engel (1971) and Fang et al. (2015), is also reproduced by the model proposed in this work.

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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 the experiments measuring the flame speed under the effect of an electric field. Among the data published, we can find slight flame deceleration [7–10], slight flame acceleration [11–15] and strong flame acceleration, of up to 200% in some cases [2,4,16–18].

Three different mechanisms have been postulated to explain the effect of the electric field on flames: ionic wind, kinetic enhance-

ment by non-thermal electrons and ohmic heating. Body forces (ionic wind) are, apparently, solely responsible for the observed response in diffusion and premixed flames in complex geometries [5,19–21]. The concept of ionic wind was widened by Sánchez-Sanz et al. [1] to include the effect of thermal electrons (electron temperature equal to the background gas temperature) on flame speed. According to Sánchez-Sanz et al. [1], the external field forces a preferential direction for the diffusion of ions and electrons, modifying the spatial distribution of the charged particles. The change in the reaction rate of the reactions involving these charged species is used to explain the new computed values of the flame propagation velocity.

The effect on the flame speed of non-thermal electrons (electron temperature higher than the background gas temperature) and of the heating of the background gas as a consequence of the increase of thermal energy per unit volume associated to the

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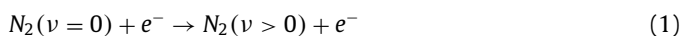
electric field has been reported as important, among others, in the microwave induced flame speed enhancement experiments carried out by [15,17,22,23] and in the computations by Bradley & Ibrahim [24] and Ju et al. [18].

Chemical effects via activation of species by high energy electrons (non-thermal) has been identified by Jagers, and Von Engel [2,11] as the main mechanism for flame acceleration in planar flames. According to Jagers, and Von Engel [2] the electrons gain energy from the field that later transfer to molecules and radicals by collisions, taking them to higher vibrational states where they are more reactive. Similarly, Van Den Boom et al. [11] indicated that excited nitrogen molecules could create an additional source of radicals in the pre-flame zone, contributing to initiate chain reactions and accelerating the oxidation process as a whole. Even when the arguments they used seem plausible, these two references simply hypothesized about a possible chemical effect but they did not provide any specific information about how the activated species could modify the chemical paths.

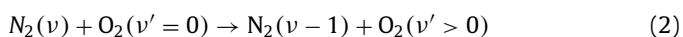
In order to identify the relative importance of each of these three mechanisms, we consider in this paper a one-dimensional model of a lean premixed flame under the influence of a longitudinal electric field. The electric field is oriented in the direction of the gas flow and can have positive or negative 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 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 [25], we chose the two-step, chain-branching chemistry model developed originally by Zeldovich [26,27] 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 et al. [28] 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 Eq. (4). Such high energy electrons might produce active radicals and electronically and vibrationally excited molecules (e.g., $N_2(v)$). According to [29], 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 III. 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 [30] or raise argon to its first electronic state [31], 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.

Symbols		Greek letters	
A_I, \dots, A_{IV}	Frequency factor of reaction I..IV	β	Zeldovich number
A, B, \mathcal{F}	Non-dimensional frequency factors	δ	Flame
C	Reaction VI multiplying factor	ϵ	Permittivity
C	Electron velocity	γ	Heat release parameter
D_T, D	Thermal/mass diffusivity	ζ	Collision frequency
E	Electric Field	Λ	Chemical enhance parameter
E_i, E_0	Autoinduced/External electric field	μ, μ_0	Eigenvalue with and without electric field
E_I	Activation energy reaction	ν	Mobility
f^0	Electron energy density function	ρ	Gas density
J	Current flux	Ω, ω	Dimensional/non-dimensional reaction rate
K_B	Boltzmann constant		
Le	Lewis number		
m	Mass		
n	Number density		
q, Q	Dimensional/Non-dimensional heat of reaction	Subindex	
		e	Electron
R_g	Ideal gas constant	F	Fuel
S_L	Flame speed	h, H	Heavy particle
T	Temperature	Z	Radical
T_c, T_0	Branching/ambient temperature	Z^+	Positive ion
u	Displacement velocity		
$(\bar{W})W$	(Mean) Molecular weight	Superindex	
x	Spatial coordinate	$'$	Dimensional quantity
Y	Mass fraction		

Compared with air at the same temperature, the high-temperature reaction zone of a hydrocarbon flame exhibits an unusually high degree of ionization as a consequence of the chemi-ionization reactions. It usually depends on the fuel burned, but it is generally accepted that the flame constitutes a weakly ionized, collisional plasma in which the concentration of electrons is much smaller than the concentration of neutral particles [32,33]. Most of the previous modeling efforts considered thermal electrons (electron temperature equal to the background gas temperature) [1,20,34], 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 [29]. 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

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