Electric Field Assisted Reduction of NO\textsubscript{x} Emission: A Numerical Study

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Abstract: The paper reports simulation results on the influence of a DC driven radial electric field on the emission characteristics; especially NO\textsubscript{x} and CO of a premixed methane/air laminar jet flame. A multi-physics computational model is developed in the OpenFOAM framework to simulate electric field coupled premixed combustion process. The model predictions show that for an applied voltage of 50 kV, the flame structure changes significantly for both stoichiometric and fuel rich conditions. For the fuel rich condition, the ionic wind allows additional mixing of the fuel rich stream with the surrounding air and drastically altering the flame structure. The electric field was found to reduce the NO\textsubscript{x} emission significantly. Over the entire range of flowrate conditions, the stoichiometric fuel-oxidizer mixture showed a decrease in maximum NO\textsubscript{x} in presence of electric field. For the fuel rich case, however as the flow rate is increased the NO\textsubscript{x} reduction factor decreased from 12.0 to 1.6. For CO emissions, the presence of electric field reduces the concentration under fuel rich conditions and vice versa for the stoichiometric flame. The role of kinetics is analyzed and discussed.

Keywords: Chemi-ionization, Electric field, Ionic wind, NO\textsubscript{x} kinetics

1. Introduction
For several decades, researchers have been interested in the electrical properties of flames and how they can be controlled by the application of electric fields. It has been shown that electric fields affect flames and combustion processes in three distinctive and major ways – thermal effect [1], ionic wind effect [2] and electro-chemical effect [3, 4]. The thermal effect contributes to the neutral gas heating through Joule heating when there is large current across the electric field. The ionic wind effect causes fluid dynamic changes in the flow field via electrical body force resulting from space charge and electric field. The electro-chemical effect produces energetic electrons, ions, radicals and excited molecules in the gas stream which directly contributes to the reaction kinetics.

Studies involving experimental and modeling approaches have been conducted, in an effort to elucidate the influence of both direct-current (DC) and alternating-current (AC) electric fields on the combustion characteristics. Experimental studies have demonstrated that DC electric fields have a strong influence on the flame shape [5], flame propagation speed [2], emission and soot characteristics [6, 7]. Simulations employing multi-physics models have shown the role of ionic wind under DC external fields [8-10]. In recent years, there has been growing interest in utilizing AC electric field to modify the combustion characteristics of flames [11, 12]. Despite the large volume of work on electric field – flame interactions, studies related to the impact of electric
field on NO\textsubscript{x} emission is limited. Vatazhin et al. [13] in their laminar propane diffusion flame experiments observed up to 30% reduction in NO\textsubscript{x} emission with respect to the emission index with the implementation of electric field on a negatively polarized burner. Zake et al. [14] applied a DC electric field in a flame channel flow and observed a reduction in the NO\textsubscript{x} emission by \sim 80%. The decrease in the emission characteristics was attributed to the reduction in the flame temperature in presence of the electric field. Sakhrieh et al. [6] experimentally investigated the influence of electric field on premixed methane/air flames at elevated pressures. They observed as much as 95% reduction of CO emission, accompanied by 25% increase of NO\textsubscript{x} irrespective of pressure. They attributed the decrease in CO to ionic winds that changed the flame geometry and reduced the amount of incompletely burned fuel. However, explanation related to the increase in NO\textsubscript{x} was not provided. In their experimental work, Vega et al. [5] showed that for a premixed CH\textsubscript{4}/O\textsubscript{2}/N\textsubscript{2} flame, the NO\textsubscript{x} emission is unaffected under applied electric field conditions for which the flame remains undeformed. In a recent paper, Zhang et al. [12] examined the behavior of NO emission of laminar non-premixed CH\textsubscript{4}/air flame when subjected to high frequency (10 kHz) AC electric fields. Over the voltage range of 0 – 4.0 kV, a non-linear response of NO emission was observed. In between 0 – 1 kV peak voltage, the NO in flue gas showed a sharp decrease which then steadily increased to high values for 1.0 – 3.0 kV, followed by a steady decrease by further increase of the applied peak potential to 4 kV.

In this paper, we numerically investigate the effects of an externally driven radial DC electric field on the flame and combustion characteristics, more specifically the NO\textsubscript{x} and CO emission in a laminar premixed CH\textsubscript{4}/air jet flame under atmospheric condition. The simulation was conducted for a multi-dimensional configuration representative of a laboratory scale mockup of an industrial system at ClearSign Combustion Corporation. A range of equivalence ratio 3 are investigated. The flame structure is analyzed, and the role of underlying thermo-kinetic/transport properties on emission characteristics is elucidated. The kinetic analysis shows that the NO\textsubscript{x} recycling pathways undergo distinct changes in presence of the external fields. The predicted NO\textsubscript{x} and CO concentration are compared against experimental measurements and are found to be in favorable agreement.

2. Experimental Setup and Procedure
A custom test furnace was designed and built to study the effects of electric fields on the combustion process. The furnace was designed to operate up to 5.86 kW at temperatures up to 1366 K. As shown in Fig. 1a, the furnace consists of a main vessel, a transition, a stack, and a burner assembly (not shown in figure). The main vessel sits on four standoff electrical insulators capable of withstanding up to 50 kV. The support stand is kept at ground potential while the rest of the vessel is electrically floating. The main vessel has two gated 0.15 m quartz windows and four 0.051 m viewing ports all on the same plane. There is a total of 26 K-type thermocouples located throughout the furnace to measure the temperatures of vessel wall and axis, cooling jackets and the top and bottom of the stack. Fuel and air are monitored using FMA 2300 series omega mass flow meters. NO\textsubscript{x}, CO and CO\textsubscript{2} emission analyzers allow assessment of emission in the stacks.

3. Numerical Modeling
The numerical study is performed using a multi-dimensional, reacting flow computational code that has been developed in the OpenFOAM framework, the details of which (both the physics
and the numerical schemes) have been thoroughly presented in [15]. In brief, the mathematical model comprises of time dependent conservation equations of total mass, species mass fraction, mixture momentum, and mixture energy, together with a Poisson’s equation to resolve the electric field distribution. The momentum conservation includes the effect of electrical body force i.e. \( q \vec{E} \). Conservation equation for both electrons and ions are solved including the effect of drift velocity \( v_{\text{drift}} = \mu_{\text{mobility}} \vec{E} \) of the ionic species. Even though electrons are included in the computation, their effect on the system is not significant due to their very low number density and extremely high mobility.

The hydrocarbon/\( \text{NO}_x \) model of Ahmed et al. [16] consisting of 301 species and 1945 reactions is used as the chemical kinetic model for neutral species, culminated from the previous works of the authors [17]. An eleven-step ionic reaction mechanism and associated thermodynamics and transport property from [8, 18] is appended to the hydrocarbon/\( \text{NO}_x \) model. The ionic mechanism consists of 6 species (electrons, \( \text{HCO}^+ \), \( \text{H}_2\text{O}^+ \), \( \text{C}_2\text{H}_3\text{O}^+ \), \( \text{CH}_3^+ \) and \( \text{C}_3\text{H}_3^+ \)). The electron mobility values are obtained from Sakhrieh et al. [6] and the Einstein relationship \( D_e = \mu_e k_B T / q_e \) is used to determine the diffusivity from the mobility values.

A schematic of the experimental test scale setup at ClearSign Combustion Corporation and the computational domain are presented in Fig. 1b. The domain takes into account only the burner and part of the post-combustion zone of the experimental setup. The domain contains part of the burner height (i.e. 0.15 m) and covers a 1.0 m long post-combustion zone having a constant diameter of 0.6 m. To reduce computational overhead, the simulations are performed on a 2-D axisymmetric configuration with structured non-uniform mesh. All the simulations reported here are for a domain composed of 10850 mesh elements for which grid independent results were confirmed. The burner acts as the powered electrode (50 kV for base case simulations) with the side walls grounded (0 kV). Both are prescribed with an isothermal (i.e. 300 K) boundary conditions. The side walls and the burner surfaces are considered to be reactively non-participating for the neutral the neutrals species, but the ionic species reaching the surfaces are
prescribed to undergo quenching/neutralization reactions [19]. A Dirichlet boundary condition for velocity is employed at the inlet, representing the different flow rate conditions considered. Outflow boundary conditions are provided at the outlet of the tubular section. Premixed methane/air mixtures at three different equivalence ratios (\(\phi = 0.5, 1.0 \) and 3.0) and flowrates (~3.45, 6.70 and 9.93, representative Reynold’s number of ~376, 728, 1081) conditions are simulated. An initial high temperature region, of 2100 K is prescribed to ensure ignition of the fuel/air mixture. All simulations are conducted for an operating pressure of one atmosphere.

4. Results and Discussion

Figure 2 presents the center line axial velocity, temperature and NO distribution for a flow rate condition of ~3.45 slpm (Re = 376) for the two different equivalence ratios of \(\phi = 1.0 \) and 3.0. These two cases are referred to as the two base case conditions. The variations due to the presence of electric field is also summarized. We would like to point out that for \(\phi = 0.50\), despite multiple simulation attempts the model always predicted an extinction of the flame with and without the presence of the external electrical field. The presence of electric field and ionic species increases the jet velocity significantly. For \(\phi = 1.0 \) and 3.0 the peak axial velocity is by a factor of ~6 and 17 respectively. The increase in the velocity results in stretching of the flame at \(\phi = 1.0 \) and a complete change of the flame structure at \(\phi = 3.0 \) (Fig. 3). Among the different ions, \(\text{H}_3\text{O}^+\) predicted to have the maximum density followed by \(\text{HCO}^+\) for the both cases. The model predicted a \(\text{H}_3\text{O}^+_{\max}\) of 8.0 x 10^{15} m^{-3} and 2.8 x 10^{15} m^{-3} and \(\text{HCO}^+_{\max}\) of 3.2 x 10^{13} m^{-3} and 3.0 x 10^{12} m^{-3} for \(\phi = 1.0 \) and 3.0 respectively. The resulting peak electrical body force is found to be 3875 N/m^3 and 1055 N/m^3 respectively.

The temperature distribution at the centerline (Fig. 2b) clearly shows a decrease in the temperature due to the ionic wind effect. The decrease in the peak temperature for \(\phi = 1.0\) is minimal, ~40 K. For the fuel rich condition, the peak temperature is lowered by ~230 K. Additionally, in presence of the electric field the peak temperature shifts closer to the burner inlet with a sharper gradient downstream. The centerline NO evolution resembles the temperature distribution. The electric field is found to significantly decrease the peak NO concentration specially under fuel rich condition.

![Figure 2. Centerline distribution of (a) axial velocity, (b) temperature, and (c) NO concentration with and without electric field for a premixed CH4/air mixture with a flow rate of 3.45 slpm, 50 kV applied voltage](image)

The spatial distribution of the OH concentration contours are compared to assess the change in the flame structure due to the electric field and the associated ionic wind effect, illustrated in Fig.
For both fuel loading the flame structure is radially constricted. An axial stretching is only observed for the stoichiometric fuel loading. The OH distribution under fuel rich conditions in absence of electric field show a distinctive flame structure. Even though premixed CH\(_4\)/air are injected the extremely rich fuel loading results in a partially-premixed fuel-oxidizer flame dynamics. Due to the entrainment of the chamber air to the fuel stream the fuel-oxidizer diffuses and mixes in radial direction and establishes a flame in the periphery. The core remains fuel rich and until sufficient oxidizer reaches the core region. Downstream of the burner inlet as fuel is depleted along the periphery additional from the surroundings gets transported into the core thereby extending the reaction zone into the core of the jet. A second peripheral reaction zone is established as seen in the OH profile which is strictly dictated by the amount of unburnt fuel available in the jet stream. The double peak in the temperature (Fig. 2b) coincides with the OH peaks in the center line. High concentration of CO overlaps with the regions of low OH in the jet core. The distribution of CH\(_4\) concentration confirms that fuel in the jet stream is either partially oxidized to CO or completely oxidized to CO\(_2\). In presence of electric field, the OH distribution for \(\phi = 3.0\) confirms that a flame structure/reaction zone close to the burner inlet is established. In addition, the OH profile looks similar to the stoichiometric fuel loading but has a smaller axial extent.

Figure 3b compares the impact of electric field on the NO\(_2\) distribution for the base cases. Under stoichiometric fuel loading and low flow rate electric field has minimal effect on the NO\(_2\) emission characteristics. The spatial distribution remains fairly unaltered with a slight increase in the maximum value; less than 4 ppm. A stark contrast is observed for the fuel rich condition both in spatial distribution and peak value of NO\(_2\). In absence of the electric field the NO\(_2\) is formed in the outer periphery of the flame due to radial gradient in the temperature and through NO-NO\(_2\) recycling reactions. The peak NO\(_2\) is decreased by a factor \(~ 20\) by the electric field. At the same time the NO\(_2\) are formed in the post combustion region downstream of the flame location.

The impact of electric field on the overall emission characteristics was assessed by comparing the peak CO and total NO\(_x\) (NO+NO\(_2\)) prediction for the different flow rate fuel loading conditions. The peak CO concentration for \(\phi = 1.0\) increases by \(~ 50\) ppm as the flow rate is increased. However, in presence of electric field a non-linear trend is apparent. The sharp transition in the maximum CO concentration occurs at the highest flow rate where a lifted flame is established. The lifted flame structure increases the region of incomplete combustion and
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results in the increase in CO formation in the domain. In the lower flow rate range the CO concentration increases slightly due to the fact that the radial constriction of the flame increases the gap between the burner rim (i.e. nozzle diameter) allowing some of the fuel-air mixture to bypass the reaction zone in the core. A similar behavior was reported in [6]. In contrast, the rich case shows an opposite trend where CO is reduced at the maximum at the lowest flow rate. However, over the entire flow rate range, the electric field is found to be effective in reducing the CO emission; allowing complete combustion to occur by mixing of the fuel rich jet stream with ambient air. The total NO\textsubscript{x} under stoichiometric fuel loading decreases in a linear fashion and the electric field consistently decreases the maximum total NO\textsubscript{x} by a factor of \sim 1.6. For higher fuel loading the total NO\textsubscript{x} is reduced by the electric field but the emission increases with flow rate. At $\phi = 3.0$ an increase in the flow rate results in a higher flame temperature, contributing to increase NO\textsubscript{x} production.

The influence of applied electric on the kinetics involved in NO\textsubscript{x} formation, and NO-NO\textsubscript{2} interconversion was assessed by comparing the variation in the rate of formation of NO and NO\textsubscript{2} through the different reactions. For this purpose, two different regions in the computational domain (Zone1: 0.17 – 0.27 m and Zone2: 0.55 – 0.65 m above the burner surface), are selected, which represents the location of highest temperature gradients. The rates of production of NO and NO\textsubscript{2} by the individual reactions are volume integrated in these regions and normalized against the case without the electric field. The comparisons for stoichiometric condition are shown in Fig. 5. The decrease in flame temperature due to the application of electric field in zone 1 makes the extended Zeldovich channel (N\textsubscript{2} + O = NO + N, N + O\textsubscript{2} = NO + O, N + OH = NO + H) insignificant in that region. Instead, the direct NO formation channel from the recycling reaction NO\textsubscript{2} + H = NO + OH becomes important. Besides, most of the NO in this low temperature condition in presence of electric field reacts with fuel fragments to form stable intermediates HCN and HCO. However, with the absence of electric field, most of the NO participates in direct NO-NO\textsubscript{2} or NO-HNO interconversion reactions.

For zone 2 downstream of the domain, where the temperature gradients are different from those at zone 1, a significantly different set of NO\textsubscript{x} formation and recycling reactions are observed. The applied electric field changes the major NO formation path from direct oxidation (NO\textsubscript{2} +
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\[ \text{OH} = \text{NO} + \text{HO}_2 \] to reactions with atomic hydrogen \((\text{NO}_2 + \text{H} = \text{NO} + \text{OH})\). Besides, it prompts the \(\text{N}_2\text{O}\) formation channel that is absent without ion kinetics.

![Graph](image)

**Figure 5.** The significant \(\text{NO}_x\) formation and recycling reactions with and without the application of electric field at zone 1 (2 cm above the burner) and zone 2 (40 cm above the burner) for stoichiometric \((\Phi = 1.0)\) case with a flow rate of 3.45 slpm

5. **Conclusions**

A multi-dimensional Multiphysics model has been developed to simulate electric-field assisted combustion process in a self-consistent fashion. The model was employed to simulate a test scale burner setup having premixed CH\(_4\)/air mixture with detailed fuel-\(\text{NO}_x\) kinetics with an ionic reaction scheme appended to it. Simulations were conducted over a range of flow rate and fuel loading condition with an applied electric potential of 50 kV to generate radial electric-fields. The emission characteristics of two major pollutant classes- CO and NO\(_x\) are investigated. The predictions show that ionic wind effects resulting from the electric-field significantly increases the jet velocity and constricts the flame/reaction region both in the radial and axial directions. Lifted flames are also observed at limited cases. The flame constriction has a strong dependence on the fuel loading and flow rate conditions. Under fuel rich conditions, the electric-field drastically changes the flame structure by allowing mixing of the fuel stream with the surrounding oxidizing environment. A significant decrease in total NO\(_x\) is found to occur over the parametric space considered; attributed to a decrease in the flame temperature. Under fuel rich conditions, the electric-field is found to decrease the CO emission but vice versa for stoichiometric condition. The radial constriction of the flame under stoichiometric fuel loading allows unburnt fuel to bypass the core reaction regime. Kinetics analysis indicates that in presence of electric-field, the NO formation route shifts from Zeldovich to direct NO formation through \(\text{NO}_x\) recycling reaction and forms stable intermediates.
6. References


