Modeling ion and electron profiles in methane-oxygen counterflow diffusion flames

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Abstract

Charged species formed in a laminar counterflow diffusion flame of methane and oxygen enriched air are studied numerically. Mole fractions of major individual charged species are predicted using a one-dimensional counterflow diffusion flame model. The Cantera software and the laminar counterflow flame model is used to solve one-dimensional conservation equations of reactive flows to obtain the spatial profiles of velocity, the mole fractions and the temperature in the steady state. The distance between the two nozzles is kept at 2 cm. A detailed transport model and a 65 step mechanism involving 11 charged species in addition to the 208 step methane-air combustion mechanism with neutral species is used to model the electrical properties of the flame. Strain rate is changed from 10s\(^{-1}\) to 90s\(^{-1}\) in steps of 10s\(^{-1}\) by adjusting the mass fluxes of the fuel and oxidizer. The oxygen content in the oxidizer is varied from 21% to 100%. The effects of strain rate and oxygen content variation on the charged species profiles are comparatively analyzed. Electrons and H\(_3\)O\(^+\) appear to be the most dominant negative and positive charged particles respectively. OH\(^-\) is found to be the most dominant species amongst the negative ions. The increase in oxygen content from 21% to 100% causes approximately twofold increase in both the electron and H\(_3\)O\(^+\) concentrations. The maximum concentration of electrons increases from 1.4\times10^{11} \text{ cm}^{-3} to 2.8\times10^{11} \text{ cm}^{-3} by increasing oxygen content in the oxidizer from 21% to 100% respectively. Also the maximum concentration of H\(_3\)O\(^+\) increases from 1.8\times10^{10} \text{ cm}^{-3} to 2.4\times10^{10} \text{ cm}^{-3} by increasing oxygen content in the oxidizer from 21% to 100% respectively. The maximum concentration of OH\(^-\) increases from 10^9 \text{ cm}^{-3} to 7\times10^9 \text{ cm}^{-3}. The total negative charged species excluding the electrons as well the total positive ions are computed and it is shown that the amount of negative ions is negligible as compared to the positive ions and the electrons are the most dominating negatively charged particles. The computational results obtained through this solution need to be verified with experimental data.

1. Introduction

Combustion is a very complex system due to the involved physical and chemical processes. During these processes many species are produced. Amongst them are the charged species, i.e. ions and electrons. Even though they are present in small amounts they play a vital role in many reactions and they can be used to our advantage for numerous applications. The mechanism by
which the ions are produced is not completely understood. The charged species play a role in certain reaction pathways which are responsible for pollutant formation like soot as found by Calcote et al. (1988). As found by Homann et al. (1988) Certain unobservable neutral species in soot formation follow the profiles of ionic species, so studying charged species can help us understand the behavior of neutral species formed in a flame. There also has been a strong interest in understanding the charged species of rocket exhaust which has directly been linked with the charged species of a flame as mentioned in the review by Fialkov (1997). It has been well known that a flame can be affected by an external electrical field which in turn can help in altering the combustion processes due to the ionic wind effects. Electrical conductivity can be measured because of the presence of charged species which provides useful information on the combustion processes [4]. Experimental studies on charged species in laminar premixed flames have been performed extensively in the past and have been reviewed comprehensively by Fialkov [3]. Even though experimental studies are of a great importance it is vital to understand the charged species formation and properties of a flame by performing numerical simulations. Unfortunately not a lot of numerical studies have been performed in this area. Recently ionic species and electrons in a laminar premixed methane oxygen flame burning with a stoichiometric ratio, $\Phi = 0.216$ at atmospheric pressure were studied comprehensively by Prager et al. (1997). The purpose of the study was to validate the experimental results obtained by Goodings et al. (1979). The simulations have produced results which match the experimental results with reasonable errors.

Many of the actual combustion processes involve diffusion flames but unfortunately not many studies have been performed that predict the ion and electron concentrations in a diffusion flame. In the present study, simulations of ionic species and electrons in a laminar diffusion counterflow flame burning at atmospheric pressure are performed.

2. Numerical Method

2.1 Model Description
The open source code, Cantera developed by Goodwin (2009), and the laminar counterflow flame model is used to solve one-dimensional conservation equations of reactive flows to obtain the spatial profiles of velocity, the mole fractions and the temperature in the steady state.

2.2 Reaction Mechanism
The reaction mechanism used for methane-oxygen combustion is a 208 step reaction involving 38 neutral species developed by Warnatz et al. (1997). The same mechanism was used to simulate the experimental results of Goodings et al. (1979) which produced reasonable results. This mechanism describes the chemistry of the combustion very well because it has been validated in a number of shock-tube experiments as mentioned by Prager et al. (2007). For our interest a 65 step reaction involving 11 charged species and chemi-ionization reactions and other reactions involving charged species are included as studied by Prager et al. (2007). The major positive charged species or the cations considered are $\text{CHO}^+$, $\text{H}_2\text{O}^+$, $\text{C}_2\text{H}_3\text{O}^+$ and $\text{CH}_3\text{O}^+$. The negative charged species used are electrons, i.e. $\text{E}^-$ and anions $\text{O}_2^-$, $\text{OH}^-$, $\text{O}^-$, $\text{CHO}_2^-$, $\text{CHO}_3^-$ and $\text{CO}_3^-$.
2.3 Input Data
The input data consists of three types, i.e., thermodynamic, transport and the reaction mechanism. The thermodynamic data was the one used by Prager et al. (2007) for the simulations of methane-oxygen laminar premixed flame. For transport properties a standard database as compiled by Kee et al. (1983) is used. For few species whose transport data was unavailable the values of the available transport properties of the species similar in molecular weight were used. The transport coefficients such as the species diffusivity and heat conductivity in the flame are calculated by Cantera using the Chapman-Enskog theory. Ambipolarity of diffusion fluxes which might influence the distribution of the charged species are not accounted in this study.

3. Results & Discussion
Our simulations consider a counterflow diffusion flame formed by methane and oxygen enriched air. The fuel nozzle is on the left hand side and that of the oxidizer is on the right. The effects of strain rate and oxygen content on the charged species profiles are studied. The strain rate is varied from 10 s$^{-1}$ to 90 s$^{-1}$ in steps of 10 s$^{-1}$ by adjusting the mass fluxes of the fuel and oxidizer and the distance between the two nozzles is kept fixed at 2 cm and the oxygen content for this study is kept constant at 21%. Then the effect of oxygen content is studied by changing the oxygen concentration from 21% to 100% by keeping the strain rate constant at 20 s$^{-1}$.

![Fig. 1. Mole fraction of major charged species at 21% O$_2$ and strain rate of 10 s$^{-1}$](image1)

![Fig. 2. Mole fraction of major charged species at 21% O$_2$ and strain rate of 50 s$^{-1}$](image2)

3.1 Mole Fraction
3.1.1 Effect of strain rate
Figures 1 and 2 are logarithmic plots of mole fractions of major charged species as a function of distance between the two nozzles at increasing strain rate. The maximum mole fraction of
electron increases by a very small amount which is about 12%. The maximum mole fraction of H$_3$O$^+$ cation increases from $5.0 \times 10^{-9}$ at a strain rate of $10$ s$^{-1}$ to approximately $1.2 \times 10^{-8}$ at a strain rate of $90$ s$^{-1}$ and that of OH$^-$ ion increases from $2.2 \times 10^{-10}$ to $4.0 \times 10^{-10}$.

3.1.2 Effect of oxygen content

Figure 1 and Figs. 2, 3 are logarithmic plots of mole fractions of major charged species as a function of distance between the two nozzles at increasing oxygen content in the oxidizer. The maximum mole fraction of electron increases from $4.0 \times 10^{-8}$ to $8.0 \times 10^{-8}$ and that of H$_3$O$^+$.
increases from $5.0 \cdot 10^9$ to $9.0 \cdot 10^9$ by increasing oxygen content in the air from 21% to 100% respectively. The OH ion increases from $2.8 \cdot 10^{10}$ to $2.56 \cdot 10^9$ with the increase in oxygen content.

3.2 Species Concentration

3.2.1 Effect of strain rate

Figs. 5, 6 are logarithmic plots of concentration or also known as number density of major charged species as a function of distance between the two nozzles at increasing strain rate. The maximum number density of electron increases by a very small amount from $1.4 \cdot 10^{11}$ cm$^{-3}$ to $1.7 \cdot 10^{11}$ cm$^{-3}$. The maximum concentration of H$_3$O$^+$ ion increases from $1.8 \cdot 10^{10}$ cm$^{-3}$ at a strain rate of 10 s$^{-1}$ to approximately $4.6 \cdot 10^{10}$ cm$^{-3}$ at a strain rate of 90 s$^{-1}$ and that of OH$^-$ ion increases from $1.0 \cdot 10^9$ cm$^{-3}$ to $7 \cdot 10^9$ cm$^{-3}$. The maxima of C$_2$H$_3$O$^+$ ion increases from $3.0 \cdot 10^9$ cm$^{-3}$ to $3.2 \cdot 10^9$ cm$^{-3}$.

![Graph 1](image1.png)

**Fig. 7.** Concentration of major charged species at 50% O$_2$ and strain rate of 10 s$^{-1}$

![Graph 2](image2.png)

**Fig. 8.** Concentration of major charged species at 75% O$_2$ and strain rate of 10 s$^{-1}$

3.2.2 Effect of oxygen content

Fig. 5 and Figs. 7 and 8 show the major charged species concentration as a function of distance for increasing oxygen in the air. From the figures it can be seen that as the oxygen content increases the number of charged species formed also increases. The maximum concentration of electrons increases from $1.4 \cdot 10^{11}$ cm$^{-3}$ to $2.8 \cdot 10^{11}$ cm$^{-3}$, the maximum concentration of H$_3$O$^+$ increases from $1.8 \cdot 10^{10}$ cm$^{-3}$ to $2.4 \cdot 10^{10}$ cm$^{-3}$ and that of OH$^-$ increases from $10^9$ cm$^{-3}$ to $7 \cdot 10^9$ cm$^{-3}$ with an increase of the oxygen content in the oxidizer from 21% to 100%.

As the oxygen content is increased the concentration peaks move towards the fuel side. The reason is the O$_2$ which is heavier as compared to N$_2$ due to which the flame gets pushed towards the fuel side as O$_2$ is increased. Electron concentration gets moderately affected by the increasing strain rate as compared to its heavier counterparts, i.e. H$_3$O$^+$, C$_2$H$_3$O$^+$ and OH$^-$. H$_3$O$^+$
increases by more than twice if the strain rate is increased from 10 s\(^{-1}\) to 90s\(^{-1}\). This does not violate charge conservation because the number of negative ions also increase dramatically unlike the effect on electrons. Increasing strain rate results in a moderate production of free electrons but produces a large amount of ions. Unlike the strain rate, oxygen content in the air has a huge impact on the electron concentration. Increasing oxygen content from 21% to 100% increases the electron concentration by two times. H\(_3\)O\(^+\) ion concentration also rises by almost two times. OH\(^-\) ion concentration increases by about 7 times.

4. Conclusion

Formation of charged species in a methane-oxygen counterflow diffusion flame was studied numerically. A reaction mechanism was used, i.e. the one chosen in the previous work on premixed flames by Prager et al. (2007). The effect of strain rate and oxygen content in the oxidizer on the species concentration were simulated. It was shown that the most dominant negatively charged particle was the electron and the positively charged particle was the H\(_3\)O\(^+\) ion. The concentration and the mole fraction of the species E\(^-\) and H\(_3\)O\(^+\) increased by about two times as the oxygen content was varied from 21% to 100%. The strain rate did not have a big impact on electron concentration as compared to the increasing oxygen content in the oxidizer, but it had a considerable effect on the major ions, i.e. H\(_3\)O\(^+\), C\(_2\)H\(_3\)O\(^+\) and OH\(^-\).

Since no ambipolar diffusion fluxes were used it can be concluded that the study is incomplete and the numerical study should be further investigated by using ambipolar diffusion fluxes in the transport model which might influence the distribution considerably and then validate the results by performing suitable experiments.

References