

Soot and NO Formation in Counterflow Ethylene/Oxygen/Nitrogen Diffusion Flames

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Introduction

Understanding the mechanisms of soot and NO_x formation in combustion process is of great interest to combustion scientists. Although many researchers have studied the formation mechanisms of soot and NO_x in various flames, fewer have dealt with the interaction of soot and NO_x processes. It is the objective of this paper to numerically investigate the soot and NO emission in counterflow ethylene diffusion flames, with the emphasis on the interaction of soot and NO processes.

Numerical Model

The flames studied are atmospheric counterflow, axi-symmetric laminar diffusion flames, with fuel issuing from one nozzle, and the mixture of oxygen and nitrogen from the opposite nozzle. The potential flow boundary conditions were employed. The radiative heat transfer from the flames was calculated by an optically thin model. Soot nucleation, growth and oxidation in the flames were described by an acetylene based semi-empirical two-equation soot model [1].

The discretized governing equations were solved by Newton's method. Adaptive refinement of meshes was done. The chemical reaction mechanism used is essentially from GRI-Mech 3.0. The thermal and transport properties were obtained by using the database of GRI-Mech 3.0.

Results and Discussions

Three flames were calculated. The fuel stream is pure ethylene for all the three flames, while the oxidant streams are different mixtures of nitrogen and oxygen. The mole fractions of oxygen, called oxygen index (X_{O_2}), of the oxidant stream for the three flames are, respectively, 0.20, 0.24 and 0.28.

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The calculations were first conducted by two reaction schemes: the full GRI-Mech 3.0 and its revised version obtained by simply removing all the reactions and species related to NO_x formation. It was found that there is almost no recognized difference in the predicted soot volume fraction profiles between the results obtained by the two reaction schemes for all the three flames. Therefore the formation of NO_x almost does not affect the soot production and oxidation in these flames. Consequently the rest of the paper concentrates on the influence of soot on NO emission. All the results hereafter were obtained by the full GRI-Mech 3.0 reaction scheme.

The predicted soot volume fractions are compared with those measured by [2] in Fig. 1. The global axial velocity gradient in the experiment is the same as the stretch rate (27.5 1/s) in the simulations. The stagnation planes of both experiment and simulations are at $X = 0.0$ cm in all the plots. The fuel stream comes from the right side, and the oxidant stream from the left side. It is observed that the simulation captured the general soot characteristics in these flames.

Soot may affect NO emission through the variations of radiation induced temperatures and the reaction induced radical concentrations, i.e. effects of thermal and chemical reaction. To identify these effects, three simulations were conducted for every flame. The first simulation (SIM1) was conducted by including both NO_x and soot in the model, while the inception and surface growth rates of soot were set as zero in the second (SIM2) and third (SIM3) simulations for every flame. In addition, the temperatures of SIM2 were kept the same as those in SIM1, and were calculated in SIM3. Therefore the result difference between SIM1 and SIM2 is mainly caused by chemical reaction effect of soot, while the difference between SIM2 and SIM3 is mainly caused by the radiation induced thermal (temperature) effect.

Soot does affect NO formation, and the influence is enhanced when the oxygen index in the oxidant stream is increased, as shown in Figs. 2 and 3. This is apparently due to the increase of soot volume fraction in the flame when X_{O_2} is increased.

Moreover, Figs. 2 and 3 show that the influence of soot on NO formation is mainly through chemical reaction, when X_{O_2} is 0.20 (more clearly shown in the inset of Fig. 3). However, with the increase of X_{O_2} , the radiation induced thermal effect increases. For the flame with X_{O_2} of 0.28, thermal effect becomes more significant than chemical effect. The variation of the relative effects of thermal and chemical reaction of soot on NO formation is caused by disparities in soot volume fractions and the relative contribution of thermal and prompt routes to the total NO production in the flames. The simulations indicate that when X_{O_2} is 0.20, most NO in the flame is formed

through prompt route, while the thermal route contribution to NO formation is increased with the increase of X_{O_2} , as shown in Fig. 4 by the rates of the initial thermal route reaction and the main prompt route reaction.

For the flame with X_{O_2} of 0.2, the soot volume fraction in the flame is lower. Consequently radiation induced temperature variation due to soot is negligible, as shown in Fig.5, and thus the thermal influence of soot process on NO is very small. However, the chemical process of soot causes the variation in the concentration of radical CH in the flame, as shown in Fig. 6, in spite of the lower soot volume fraction. This variation results in the disparity in the reaction rates of the most important initial reaction, $CH + N_2 = HCN + N$, of prompt route for NO formation. Accordingly soot affects NO formation in this flame mainly through chemical reaction, since prompt route dominates the production of NO in this flame and soot affects the prompt route initial reaction $CH + N_2 = HCN + N$ through the variation in the concentration of radical CH.

When X_{O_2} is increased to 0.28, the contribution of thermal route to NO formation increases due to the flame temperature rise, Fig. 4. Because of the higher soot volume fraction in this flame, the soot induced temperature variation increases, compared to the flame with X_{O_2} of 0.20, Fig. 5. Meanwhile the variations in the concentrations of radicals O and OH due to temperature effect also increase. These variations cause the influence of thermal effect of soot on NO formation to increase. Although the high soot volume in this flame also results in the variations in the concentration of radical CH and the reaction rate of the prompt initial reaction, this effect is smaller than the thermal effect. Consequently the effect of soot on the NO emission in the flame with X_{O_2} of 0.28 is mainly through the thermal effect.

Conclusions

The NO emission has almost no effect on soot in counterflow ethylene diffusion flames. When the oxygen index of the oxidant stream is lower, the relative influence of chemical reaction caused by soot production on NO formation is more important, while the relative influence of radiation induced thermal effect becomes dominant for the flame with higher oxygen index in the oxidant stream. The variation of the relative influence of thermal and chemical reaction is caused by the variations in soot volume fraction and the relative contribution of thermal and prompt routes to NO formation in the flame.

References

1. Guo, H., Liu, F., Smallwood, G.J., and Gülder, Ö.L., *Combust. Theory Modelling* 6: 173-187 (2002).
2. Hwang, J.Y., and Chung, S.H., *Combust. Flame* 125: 752-767 (2001).

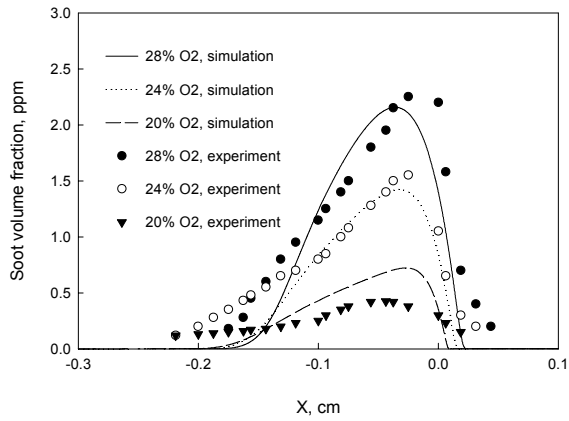


Fig. 1 Soot volume fractions obtained from the current simulations and the experiment [2].

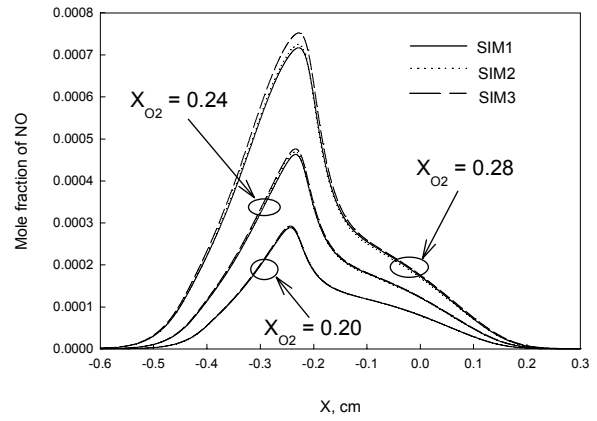


Fig.2 NO profiles of the three flames. X_{O_2} represents the oxygen index.

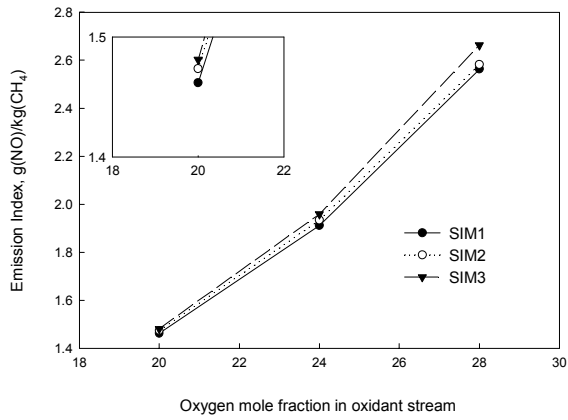


Fig. 3 Emission indices of NO.

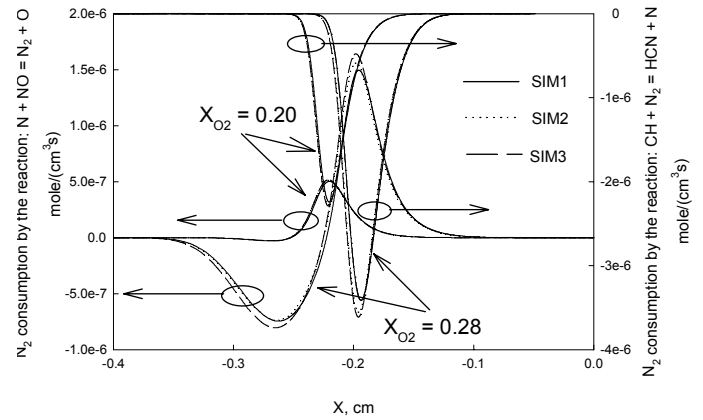


Fig. 4 Nitrogen consumption rates.

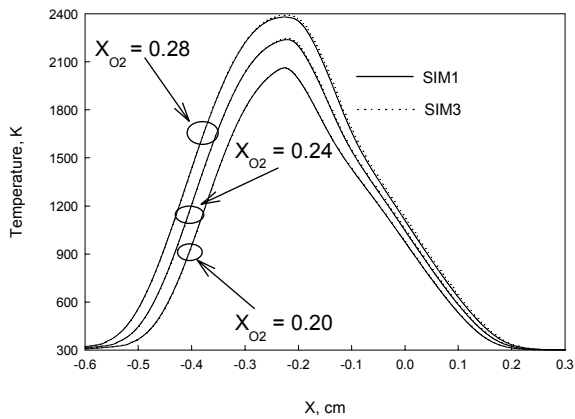


Fig. 5 Flame temperatures.

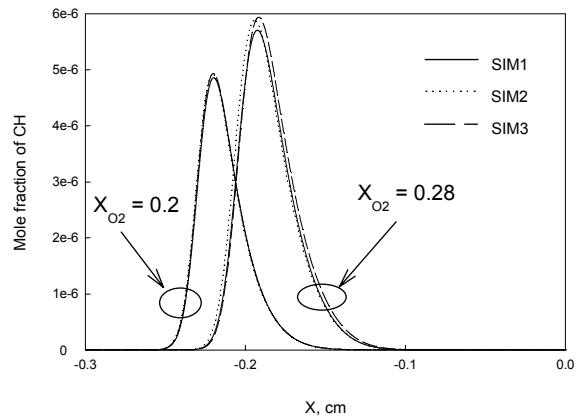


Fig. 6 Mole fractions of radical CH.