Effects of Dilution on Soot Formation in Laminar C₂H₄ Diffusion Flames

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Introduction

Improved understanding of soot formation processes in flames will undoubtedly enhance the development of effective methods of soot control in practical energy producing combustion devices. Exhaust gas re-circulation (EGR) is a widely used soot control technology. In the EGR system, exhaust gas are re-circulated into the intake manifold where it mixes with the incoming air/fuel mixture. The resulting lower combustion temperature reduces the emission levels of both soot and NOx. Therefore, there has been special interest in studying the effect of CO_2 addition on soot formation since CO_2 is a major component in the re-circulated gas mixture.

The effects of various inert additions, including CO_2 , on soot formation in non-premixed (diffusion) flames have been investigated by a few studies in the past [1-4]. Early studies only considered the qualitative nature of dilution on flame properties (such as the flame lengths) and did not measure the soot concentrations [1-2]. Later studies showed that both thermal and chemical effects were present in CO_2 diluted flames [3-4]. However, direct experimental verification of the presence of chemical effects is lacking because the effects of CO_2 addition to the species concentrations were never investigated. Motivated by the importance of understanding soot suppression in the presence of CO_2 and other gaseous additives, the specific objectives of the present study were to improve the current understanding of the soot formation processes in diluted diffusion flames and the chemical effects of CO_2 by detailed flame structure and soot concentration measurements.

Experimental Methods

The test apparatus and the measurement techniques were similar to earlier work conducted by one of the author and his coworkers and will be described only briefly, refer to references [5-7] for additional details. Four C_2H_4 flames diluted by CO_2 , N_2 , He and Ar (1:1 by volume) burning in coflowing air at atmospheric pressure were studied. The flames were supported on a coflow burner consisting of stainless steel honeycomb having 1mm cell sizes. The burner was 127 mm long and had a 35 mm diameter inner port for the fuel mixtures, and a 86mm diameter co-annular outer port to support the air coflow. The following properties were measured along the axes of the diffusion flames: soot volume fractions by laser extinction, flame temperatures by rapid insertion thermocouples with radiation correction, and concentrations of major stable gas species by Gas Chromatography, refer to references [5-7] for additional experimental details. Photographs of He and CO_2 diluted C_2H_4 diffusion flames are shown in Figures 1 and 2 for illustration.



Figure 1: Photograph of Helium diluted atmospheric laminar C_2H_4 diffusion flame.



Figure 2: Photograph of CO_2 diluted atmospheric laminar C_2H_4 diffusion flame.

Results and Discussion

Measured soot volume fractions and temperatures along the axes of C_2H_4 flames diluted by CO_2 , N_2 , He and Ar were plotted in Figures 3 and 4 as a function of height above the burner exit. The effectiveness of soot suppression increased in the order of He, Ar, N_2 and CO_2 as indicated by decreased (peak) soot volume fraction. The dilution effects were isolated from the thermal and chemical effects by using identical dilution level. Therefore, the resulting differences in the soot yields were completely due to the differences in chemical and thermal properties of these flames. Glassman [8] attributed this result to the thermal diffusivity differences among different



Figure 3: Measured soot volume fraction along the axes of C_2H_4 flames diluted by He, Ar, N_2 and CO_2 .



Figure 4: Measured flame temperatures along the axes of C_2H_4 flames diluted by He, Ar, N_2 and CO_2 .

diluents. He argued that effective soot growth time had a logarithmic variation with temperature whereas the thermal diffusivity had a direct proportionality effect, therefore larger thermal diffusivity would lead to longer growth distance/time for soot particles. Since the thermal diffusivity decreases in the order of He, Ar, N₂ and CO₂, naturally soot formation tendencies also decreases in this order.

Although the thermal effects have long been attributed to be the primary cause for the variation

of soot yields in diluted hydrocarbon diffusion flames and can explain such observed differences. Soot formation nevertheless takes place through chemical reactions involving soot growth species. Acetylene (C₂H₂) has been shown to be the primary species responsible for the soot surface growth [see 5-7, 9-10 and references Cited therein for more discussion]. There were two sources of C₂H₂ along the flame axes: local chemical production and mass transfer due to concentration gradients. Since C₂H₂ was largely formed near the high temperature reaction sheet, the amount of C_2H_2 present along the centerline is then affected by the diffusion of C_2H_2 from the flame sheet, assuming the formation of C₂H₂ near the flame sheet was equal. In reality, the amount of C_2H_2 formed near the flame sheet is determined by the maximum measured flame temperature which again decrease in the order of He, Ar, N₂ and CO₂. Clearly, exceptionally large the thermal diffusivity and mass diffusivity of He both 2000K. contribute to its large sooting tendency.



Figure 5. Calculated binary mass diffusivity between acetylene and He, Ar, N_2 and CO_2 for a temperature range between 300K and 2000K.

Major species concentrations along the axes of C_2H_4 flames diluted by N_2 and CO_2 were plotted in Figures 6 and 7. Typical of previous measurements [5-7], C_2H_4 decomposed quickly to form C_2H_2 , which was the most abundant hydrocarbon species except for the initial fuel. Figures 6 and 7 show that soot volume fractions reach maximum right after C_2H_2 concentration peaks, indicating the strong role played by C_2H_2 in soot growth. As the concentration of C_2H_2 starts to decrease, soot growth rate drops sharply. The species that is of special interest in studying the chemical effects of CO_2 addition is CO since its concentration is affected by the addition of CO_2 . Comparing the amounts of CO formed in Figures 6 and 7 clearly shows the chemical effects of the addition of CO_2 . For instance, comparing the measured concentrations of CO for the three C_2H_4 flames at a flame height of 50mm where CO_2 concentrations tend to peak, CO_2 diluted C_2H_4 flame has mole fraction of CO equal to 6.04%, while the N_2 and Ar diluted C_2H_4 flames have detected CO concentrations equal to 4.98% and 4.99%, respectively. Therefore, the addition of CO_2 increases the concentration of CO by as much as 21% comparing with the addition of N_2 and Ar. Clearly, there are strong chemical effects on soot formation due to CO_2 addition in the present test flames.



Figure 6. Measured major species composition of C_2H_4/N_2 flame as a function of flame height.

In order to identify the most important elementary reaction that is responsible for the dissociation of CO₂, computational study was performed for a counter-flow CO₂ diluted C₂H₄ flame (1:1 by volume) burning in air using the OPPDIF program in the CHEMKIN package [11]. Standard GRIMech 3.0 chemical reaction mechanism was used which contains 53 species and 325 elemental reactions. Figure 8 shows that the primary reaction that is responsible for the dissociation of CO_2 is the reaction $CO_2+H\rightarrow CO +OH$. Peak consumption was observed to occur near the flame sheet but in the fuel side of the flame.



composition of C_2H_4 / CO_2 flame as a function of flame height.



Figure 8. Production rates of CO_2 for a counter flow C_2H_4/CO_2 flame using the GRIMech 3.0 mechanism.

Conclusion

The present work found that the variation of soot yields in CO₂, N₂, Ar and He diluted C₂H₄ flames were attributed to both thermal diffusivity and mass diffusivity differences among the various diluents and the fuel. There were significant chemical effects in the presence of large amount of CO₂ addition. Addition of CO₂ results in the dissociation of CO₂ through reaction CO₂+H \rightarrow CO +OH. As a result, measured CO concentrations were higher in CO₂ diluted flame than N₂, Ar and He diluted flames. Addition of CO₂ also enhanced the formation of OH which helps oxidize the soot and its precursor, as well as decreases the H concentration which reduces the soot growth rate via the HACA soot growth mechanism [5-7, 9].

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