Combustion and Flame 166 (2016) 216–228

Contents lists available at ScienceDirect

Combustion and Flame

journal homepage: www.elsevier.com/locate/combustflame

Effects of simultaneous hydrogen enrichment and carbon dioxide dilution of fuel on soot formation in an axisymmetric coflow laminar ethylene/air diffusion flame

Mingyan Gu[a], Huaqiang Chu[a,*], Fengshan Liu[b]

[a] School of Energy and Environment, Anhui University of Technology, Ma'anshan 243002, China
[b] Measurement Science and Standards, National Research Council, Building M-9, 1200 Montreal Road, Ottawa, Ontario K1A 0R6, Canada

ARTICLE INFO

Article history:
Received 6 November 2015
Revised 21 January 2016
Accepted 22 January 2016
Available online 19 February 2016

Keywords:
Laminar coflow diffusion flame
Soot formation
Hydrogen enrichment
Carbon dioxide dilution

ABSTRACT

Effects of simultaneous hydrogen enrichment and carbon dioxide dilution to hydrocarbon fuels on soot formation are of fundamental and practical interest. Previous studies found that addition of either hydrogen or carbon dioxide to fuel reduces soot chemically in addition to the dilution effect in laminar coflow ethylene/air diffusion flames. A numerical study was carried out in this work to investigate the effects of adding hydrogen and carbon dioxide simultaneously to fuel on soot formation in an axisymmetric laminar coflow ethylene/air diffusion flame at atmospheric pressure. Numerical calculations were conducted using detailed gas-phase chemistry and thermal and transport properties. Soot inception is assumed to be the result of collision of two pyrene molecules. The subsequent particle surface growth, soot oxidation, and particle interactions are modeled by the hydrogen abstraction C_2H_2 addition (HACA) mechanism and a sectional model. Soot surface growth through condensation of pyrene was also taken into account. The flame model is able to reproduce fairly well the chemical effects of adding either hydrogen or carbon dioxide to ethylene observed experimentally in the literature. Addition of hydrogen is more effective on soot inception suppression and addition of carbon dioxide is more effective on soot surface growth suppression. The simultaneous hydrogen enrichment and carbon dioxide dilution to ethylene retains the individual soot suppression benefits of hydrogen enrichment and carbon dioxide dilution. These results suggest that the chemical interactions between hydrogen and carbon dioxide on soot formation are weak.

1. Introduction

Reduction of soot and carbon dioxide emissions from combustion systems has drawn unprecedented attention in recent years because they are the two most important contributors to global warming through absorption of solar radiation [1,2]. In addition, soot aerosols emitted from combustion devices and biomass burning have been found detrimental to urban air quality and human health [3,4]. Combustion of fossil fuels, i.e., hydrocarbons, unavoidably emits carbon dioxide and soot. Various technologies have been proposed to mitigate their emissions, such as the oxy-fuel combustion technology for CO_2 capture and storage (CCS) [5], the increased use of biofuels for CO_2 emission reduction, and flue gas recirculation (FGR) and fuel dilution for soot formation reduction. Due to the increased cost associated with the oxy-fuel combustion technology for CCS, it is unlikely to gain widespread use in the near future. FGR has been commonly used in various combustion devices to reduce soot and NO_x emissions, since it is effective and can be easily and economically implemented. The main compositions in dry FGR are N_2 and CO_2 in air-fuel combustion systems and only CO_2 in oxy-fuel combustion devices. Although the recycled CO_2 is primarily added to the oxidizer stream in oxy-fuel combustion systems, it is still highly relevant to understand how addition of CO_2 to fuel affects soot formation.

To gain fundamental insights into the effects of CO_2 addition, there have been many studies to investigate the effects of CO_2 addition on soot formation in diffusion flames at atmospheric pressure. As discussed by Liu et al. [6], addition of CO_2 could affect the flame structure and soot formation through dilution, thermal, and chemical effects. Earlier studies reached different conclusions about the mechanisms of soot formation suppression by CO_2 addition to fuel, e.g., enhanced soot burnout [7] and purely thermal [8]. More recent studies have established that CO_2 suppresses soot formation not only through dilution and thermal effects, but also through chemical effects [6,9-11]. These authors speculate that the chemical effect of CO_2 is to prompt gas phase soot precursor oxidation through enhanced OH concentrations. The experimental

* Corresponding author.
E-mail address: hqchust@163.com (H. Chu).

http://dx.doi.org/10.1016/j.combustflame.2016.01.023

measurements of Angrill et al. [12] showed that CO_2 addition to the oxidizer stream reduced soot formation, but did not affect soot oxidation. The numerical study of Liu et al. [6], in which soot formation was not included, revealed that CO_2 suppresses soot inception by lowering both temperature and acetylene concentration and enhancing the concentration of OH radical. The dominant pathway for the chemical effect of CO_2 was identified to be the reverse reaction of $CO + OH \leftrightarrow CO_2 + H$ [6]. The numerical study of Guo and Smallwood [13] conducted in a laminar coflow ethylene/air diffusion flame found that addition of CO_2 to the fuel side indeed suppresses soot formation chemically and the effect is to suppress soot inception and surface growth rates, but not soot oxidation rate speculated in several earlier studies [7,9-11]. The dominant pathway for the chemical effects of CO_2 was found once again through $CO + OH \leftarrow CO_2 + H$ [13]. The importance of H radical in soot inception and soot surface growth has long been recognized [14]. Both the numerical calculations [13] and experimental measurements [11] showed that the soot loading reduction by the chemical effect of CO_2 is significant and comparable to its thermal and dilution effects. Several recent studies have focused on the effects of CO_2 addition to fuel on soot formation in diffusion flames at elevated pressures, e.g., [15,16].

Although hydrogen is undoubtedly the cleanest energy carrier since its combustion does not produce CO_2 or soot, its widespread use in power generation and transportation faces severe challenges in terms of production and storage. However, it is practically feasible to use hydrogen as an additive to hydrocarbon fuels to mitigate CO_2 and soot emissions. Research conducted so far has shown that addition of hydrogen to hydrocarbon fuels improves the combustion performance [17–19] and reduces NO_x [20,21] and soot formation [22–26]. Tesner et al. [22] showed that addition of hydrogen or nitrogen to fuel reduces soot yield in a laminar cylindrical methane diffusion flame with hydrogen being less effective. Using a Wolfhard–Parker burner, Dearden and Long [23] investigated the effect of hydrogen and nitrogen addition to fuel on the sooting rate of laminar ethylene and propane diffusion flames. They found addition of hydrogen leads to decreased sooting rate in both flames and hydrogen is more effective than nitrogen when added to the ethylene flame. Du et al. [24] showed that H_2 addition to fuel substantially reduces the soot inception strain rate in counterflow C_2H_4, C_3H_8, and C_4H_{10} diffusion flames. The possible mechanisms through which H_2 affects soot formation are dilution, preferential diffusion, thermal, and chemical [24]. Du et al. discussed the potential chemical effect of H_2, but did not provide direct evidence. The experimental study of Gülder et al. [25] in laminar axisymmetric coflow C_2H_4, C_3H_8, and C_4H_{10} diffusion flames provided convincing evidence that H_2 addition to ethylene inhibits soot formation chemically. However, the additional chemical effect of the H_2 is absent in the C_3H_8 and C_4H_{10} flames. The numerical study of Guo et al. [26] reproduced the chemical effect of H_2 addition to ethylene on soot formation reduction found experimentally by Gülder et al. [25]. The numerical results reveal that H_2 addition to ethylene reduces soot formation chemically by decreasing the hydrogen atom concentration in the soot surface growth regions, which in turn lowers the soot surface growth rate via the reduced surface active site number density for C_2H_2 addition [27,28], and by increasing the H_2 concentrations in the soot inception regions low in the flame.

More recently, the effect of hydrogen addition to fuel on soot formation has been experimentally investigated in laminar coflow acetylene/air [29] and methane/air [30,31] diffusion flames. The potential chemical effect of hydrogen on soot formation suppression in the acetylene diffusion flame was only briefly discussed by Pandey et al. [29]. The experimental measurements of Migliorini [30] conducted in a laminar coflow methane/air diffusion flame with hydrogen and helium addition up to 40% (volumetric basis) found that addition of either hydrogen or helium to fuel reduces the soot loading. However, hydrogen is less effective than helium in soot formation suppression. These results imply that addition of hydrogen to methane prompts soot formation chemically, which is in contrast to its chemically suppressive effect on soot formation when added to ethylene [25,26]. Recently, Liu et al. [31] modeled the flames investigated by Migliorini [30] and their numerical results are in qualitative agreement with the experimental findings with regard to the relative effectiveness of hydrogen and helium addition in suppression of soot. The detailed pathways for the different chemical effects of hydrogen on soot formation in the methane and ethylene diffusion flames remain unclear.

Previous studies have focused on the effects of adding either CO_2 or H_2 to fuel on soot formation in diffusion flames. The effects of simultaneously adding both CO_2 and H_2 to the fuel stream on soot formation in diffusion flames have not been reported in the open literature. Understanding of the effects of adding both CO_2 and H_2 to fuel on soot formation is of importance and interest not only from a fundamental point of view, but also from practical perspectives, since it is highly relevant to soot formation in hydrogen enriched hydrocarbon combustion with carbon dioxide dilution. In this study, the effects of hydrogen addition to fuel with CO_2 dilution on soot formation are numerically investigated in a laminar coflow ethylene/air diffusion flame at atmospheric pressure. The objective of this study is to investigate the effectiveness of CO_2 dilution on soot formation suppression in the presence of hydrogen in ethylene. In particular, this study intends to ascertain if there is an interaction between the chemical effects of H_2 and those of CO_2 in soot formation suppression.

2. Numerical model

The flame code used in this study has been documented by Zhang [32] and described in details in several previous studies [31,33,34]. Therefore, the description of the overall flame model and various sub-models, such as radiation transfer and soot formation, is brief and only the main features are presented. Further details can be found in [32–34].

The governing equations have been described previously [26,32,35], which are the fully elliptic conservation equations of mass, momentum, energy, and species in the low Mach number limit and in axisymmetric cylindrical coordinates. It is important to retain the gravitational acceleration term in the stream-wise (vertically upward) momentum equation and the radiation source term in the energy equation in laminar diffusion flame modeling. The method of correction diffusion velocity [36] was followed to guarantee that the net diffusion flux of all species, including soot, vanishes in both the radial (r) and the stream-wise (z) directions. The interaction between the gas-phase and solid phase (soot) chemistry was considered for the relevant gaseous species involved in soot production and oxidation. The thermal diffusion associated with species transport was neglected except for the two lightest species, namely H and H_2 [26,35].

Radiation heat transfer was calculated using the discrete-ordinates method (DOM) coupled with a statistical narrow-band correlated-k (SNBCK) based wide-band model for the absorption coefficients of CO, CO_2, and H_2O [37]. The absorption coefficient of soot was approximated by the Rayleigh expression [38]. Further details of thermal radiation calculations can be found in [37,38].

The soot formation model employed in this study has been described extensively in previous studies [31–34]. The soot inception process, i.e., the physical and chemical mechanisms governing the transition from large PAH molecules to soot particles, is currently poorly understood. Although the soot inception step might involve multiple large PAH species, it has been simplified as the collision of two A4 (pyrene) molecules in the free-molecular region with a