Fuel 210 (2017) 472–481

Contents lists available at ScienceDirect

Fuel

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

Full Length Article

Influence of soot aging on soot production for laminar propane diffusion flames

J.P. Soussi[a], R. Demarco[a,*], J.L. Consalvi[b], F. Liu[c], A. Fuentes[a]

[a] Departamento de Industrias, Universidad Técnica Federico Santa María, Av. España 1680, Valparaíso, Chile
[b] Aix-Marseille Université, IUSTI/ UMR CNRS 7343, 5 rue E. Fermi, 13453 Marseille Cedex 13, France
[c] Measurement Science and Standards, National Research Council, Building M-9, 1200 Montreal Road, Ottawa, Ontario K1A 0R6, Canada

ARTICLE INFO

Keywords:
Laminar diffusion flame
Propane
Oxygen index
Soot aging effect
Radiant fraction

ABSTRACT

A numerical analysis was conducted to investigate the effect of varying the Oxygen Index (OI) of the oxidizer stream between 21 and 35% on soot production and thermal radiation emitted by laminar axisymmetric propane diffusion flames at atmospheric pressure. The extended enthalpy defect flamelet model, an acetylene/benzene-based two-equation semi-empirical soot production model, and the Full-Spectrum correlated-k radiative property model were used in the numerical simulations. The focus of this study is to demonstrate that it is important to account for the soot aging effect to correctly predict how increasing OI affects the predicted soot production. Three soot surface growth rate models were considered. The first model neglects the soot aging effect and assumes the soot surface growth rate is linearly dependent on soot surface area. The second and third models account for the soot aging effect by assuming the soot surface growth rate is proportional to the square-root of soot surface area and assuming a particle size-dependent sublinear soot surface area, respectively. The predicted flame height, soot volume fraction, radially integrated soot volume fraction and radiant fraction were compared to available experimental data. The first soot model predicted a much higher soot loading increase with increasing OI than observed experimentally. The second and third soot models improve considerably the predicted general behavior of soot loading increase with OI. Soot and combustion gases make comparable contribution to flame radiation under the conditions studied. When the soot aging effect is properly taken into account, the relatively efficient numerical code assessed in this study becomes a suitable tool for predicting soot production and thermal radiation in laminar propane diffusion flames at different OI conditions. Moreover, increasing OI of the oxidizer stream is a remarkable way to enhance the flame radiation where the correct estimation of soot production is essential to predict the radiant fraction of the flame.

1. Introduction

Since the combustion of fossil fuels was discovered, it has played a leading role in energy production and power demand of modern societies, although the associated pollution has undesirable health and environmental effects [1]. Despite the current efforts to increase the usage of renewable energy, the consumption of fossil fuels continues to grow along with energy consumption [2]. Therefore, an efficient management of this type of fuels is increasingly important at both industrial and domestic levels. This is mainly based on the pressing needs to further improve the combustion efficiency and at the same time to reduce combustion generated emissions.

Industrial flames are in general turbulent and great efforts have been made in the last few decades to develop and improve models to simulate the interaction between chemistry and turbulence reliably and efficiently. One of the most effective models in this topic has been the laminar flamelet concept [3]. This approach is based on the assumption that the behavior of a turbulent flame can be described by an ensemble of laminar flames subject to different strain rates. The rationale of this hypothesis can be understood based on the existence of similarities between the scalar distributions in laminar and turbulent flames [4]. Laminar diffusion flames are often used as a model flame configuration to gain fundamental understanding of different physical and chemical mechanisms involved in the combustion process, which is the necessary first step towards addressing industrial combustion issues. At the same time, this type of flame is ideal to apply optical diagnostics to obtain high quality experimental data for model validation and to permit numerical simulation using detailed reaction mechanisms due to its good repeatability and relatively simple flow field.

A distinct characteristic of non-premixed combustion is the formation

* Corresponding author.
E-mail address: rodrigo.demarco@usm.cl (R. Demarco).

http://dx.doi.org/10.1016/j.fuel.2017.08.086
Received 13 April 2017; Received in revised form 19 August 2017; Accepted 25 August 2017

of soot and its impact on flame properties, such as local temperature and thermal radiation [5,6]. In normal co-flow diffusion flames, like the ones analyzed in this research, soot particles are formed on the fuel side of the reaction zone, close to the high temperature regions. Then, these particles are transported downstream mainly by convection toward the tip of the flame, where they are oxidized by oxidative compounds such as O_2 and OH under high temperature conditions [7,8]. One of the important parameters that directly affects the flame structure, soot production process, and the related radiation is the oxygen concentration in the oxidizer stream [9], known as the Oxygen Index (OI). This parameter has an important influence on combustion reactions and flame temperature. As the amount of oxygen in the oxidizer stream is increased, the fuel pyrolysis process is accelerated, temperature is increased, and consequently the soot formation reactions are greatly enhanced. In addition, because of the higher amount of oxygen molecules available, the overall oxidation rates of soot and combustion in general are also significantly enhanced [10,11], leading to shortened flame heights. The dual role played by the OI in soot production enables a competition between the formation and oxidation mechanisms that needs to be studied from both technological and fundamental points of view. In fact, investigation of the OI effects is highly relevant to oxy-fuel and oxygen-enriched combustion technologies and is critical to advance our understanding of the characteristics of oxy-fuel and oxygen-enriched combustion.

From an industrial point of view, the strategy of using enriched oxygen for combustion to intensify reactions and to improve combustion efficiency is not new and has been implemented mainly within the context of oxy-fuel combustion with flue gas recirculation to reduce peak flame temperatures and pollutant emissions (both soot and NOx). The economic feasibility of oxy-fuel combustion has been discussed extensively such as in the edited books by Baukal [9] and Qi and Zhao [12]. Although oxy-fuel combustion with flue gas recirculation is likely the most promising technology for CO2 capture, it is recognized that oxygen-enriched combustion tends to enhance NOx emissions [13].

One of the gaseous hydrocarbons commonly used in industrial combustion systems, but not widely documented, is propane. Some domestic applications include the use of propane for heating homes, heating water, cooking, barbecuing and lighting. Propane is also widely used in industrial burners for air heating, fire tube boilers, steel forging, and glass processing. There has been relatively less fundamental research conducted in propane flames compared to that for methane and ethylene, although some studies have been carried out to understand the structure and combustion chemistry of propane diffusion flames [14–17]. In addition, the influence of oxygen enrichment on soot production [18] and radiative properties of propane coflow diffusion flames has been experimentally studied, demonstrating that higher radiation heat emission rate can be reached by increasing the OI of the oxidizer stream [19,20].

Despite the significant progress in our understanding of various soot formation processes in hydrocarbon flames and in soot formation model development [21], there is still a lack of robust and relatively simple soot models that perform equally well under different flame conditions or for flames fueled with different hydrocarbons. Although the soot inception step plays the bottleneck role in the overall soot formation process, it contributes negligibly to the total soot mass in comparison to the surface growth process. It has been well-known that soot particles gradually lose their surface reactivity as they become more mature [5,22–25]. This effect can be explained in terms of the decrease of active sites or defect sites on soot particle surface, and is normally called soot surface aging. It has been postulated that the soot aging effect is related not only to the local temperature but also to the residence time that the soot particles are subjected to, i.e., their thermal age [26]. However, the mechanism of soot surface aging has not been fully understood. Studies conducted so far have suggested that it is related to the carbonization/dehydrogenation processes of the soot particles [27]. At the surface of soot particles, the change in the chemical composition produces a decrease in the concentration of active $C-H$ sites available for reaction and therefore a decrease in the particle surface reactivity for growth.

Different approaches have been proposed in the literature to account for the decrease in soot surface reactivity within the context of the hydrogen abstraction acetylene addition (HACA) mechanism for soot surface growth, e.g. Appel et al. [28] and Veshkini et al. [26]. Within the framework of the semi-empirical acetylene-based two-equation soot model, Liu et al. [29] have evaluated two soot surface growth models: one is proportional to the square-root of specific soot surface area (soot surface area per unit volume) and the other is proportional to the specific soot surface area. They showed that the soot surface growth model based on the square-root of soot specific surface area performs much better in terms of the predicted soot distribution and the pressure dependence of the peak soot volume fraction over a wide range of pressures for methane-air diffusion flames. They explained the sublinear dependence (here square-root) of the soot surface growth rate on soot specific surface area in terms of the soot surface aging phenomenon mentioned above and the shielding effect due to soot particle aggregation.

In this study soot formation in laminar coflow propane diffusion flames was modeled, considering different OI in the oxidizer flow ranging from 21% to 35%. The simulations were carried out using a modified semi-empirical two-equation soot model and a detailed reaction mechanism of propane combustion. The numerical results were validated using experimental results obtained by Escudero et al. [20], analyzing flame height, soot loading and radiant fraction. Three soot surface growth models that have different functional expressions for soot surface area were used to demonstrate the importance of taking into account the soot aging effect on the predicted soot loading at different OI conditions.

The present study intends to demonstrate that the account of the soot aging is important to accurately predict soot production. In this sense, study different OI conditions put in evidence the need to consider the aging effect in order to provide accurate soot content predictions. Finally, the present study proposes a modification in order to improve the predictive capabilities of the semi-empirical soot model.

2. Numerical model

The overall continuity equation, the Navier–Stokes equations in the low Mach-number formulation, and transport equations for the mixture fraction (ξ) and the total enthalpy (h) were solved in axysimmetric cylindrical coordinates. These equations were solved using a finite volume method on a staggered grid. Steady-state solutions were reached by time marching. The ULTRASHARP scheme was applied for the convective terms while a second-order central difference scheme was used for the diffusion terms. The pressure–velocity coupling was dealt with using the Iterative PISO algorithm [30].

2.1. Combustion model

Chemical reactions were modeled by using the Extended Enthalpy Defect Flamelet Model (E-EDFM) [31], which provides the state relationships for scalars based on mixture fraction (ξ), scalar dissipation rate (χ) and an enthalpy defect parameter (X_R). The extended version of this model stands for the calculation of production rates of the soot model within the flamelet generation. These production rates are used as input for the soot transport equations described later. During the solution of the transport equations, the local values of the mixture viscosity, density, diffusion coefficient, temperature, species and soot production rates were extracted interactively from the flamelet library.

For the construction of the flamelet library a counterflow diffusion flame configuration was used, based on the OPPDIF code [32]. The code was modified in order to take into account the effect of thermal radiation into the energy equation. A strain rate ranging from about $10^{-1}\,s^{-1}$ up to the extinction value was considered. Regarding the