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Rate-ratio asymptotic analysis of the influence of addition of carbon monoxide on the structure and mechanisms of extinction of nonpremixed methane flames with comparison to experiments

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ABSTRACT

Rate-ratio asymptotic (RRA) analysis is carried out to elucidate the influence of carbon monoxide on the structure and critical conditions of extinction of nonpremixed methane flames. Steady, axisymmetric, laminar flow of two counterflowing streams toward a stagnation plane is considered. One stream, called the fuel stream is made up of a mixture of methane (CH_4) and nitrogen (N_2). The other stream, called the oxidizer stream, is a mixture of oxygen (O_2), and N_2. Carbon monoxide (CO) is added either to the oxidizer stream or to the fuel stream. Chemical reactions, represented by four global steps, are presumed to take place in a thin reaction zone. To the leading order the reactants, CH_4, O_2, and CO are completely consumed in the reaction zone. On either side of this thin reaction zone, the flow field is inert. These inert regions represent the outer structure of the flame. The outer structures provide matching conditions required for predicting the structure of the reaction zone. In the reaction zone, chemical reactions are presumed to take place in two layers—the inner layer and the oxidation layer. The scalar dissipation rate at extinction is predicted from results of the asymptotic analysis and compared with previous measurements and computational predictions using detailed chemistry. The predictions of the asymptotic analysis are found to agree well with the experimental data for CO addition to the fuel stream, and for small amounts of CO addition to the oxidizer stream. For large amounts of CO addition to the oxidizer stream, the approximations introduced in the asymptotic analysis become inaccurate. A key finding is that with increasing amounts of CO added to the oxidizer stream the scalar dissipation rate at extinction first increases and then decreases. It is attributed to changes in location of the inner layer within the reaction zone.

1. Introduction

Starting from the pioneering work of Peters and Williams [1] on rate-ratio asymptotic (RRA) analysis of premixed methane flames, many studies have addressed fundamental aspects of the structure of premixed and nonpremixed flames using RRA analysis [2–15]. These analyses employed reduced mechanisms that were obtained employing a systematic procedure developed by Peters [3]. The present RRA analysis is focused on elucidating the influence of carbon monoxide (CO) on the structure and extinction of nonpremixed methane flames.

Studies on combustion of mixtures made up of several reactants are of fundamental and practical importance. The practical importance of these studies emanates from a need to characterize combustion processes taking place in practice where the fuel is made up of several components that include methane and carbon monoxide. The present investigation is motivated by a previous experimental and computational study of the influence of carbon monoxide (CO) on the structure and critical conditions of extinction of nonpremixed methane (CH_4) flames [16]. In this previous study, a Burke-Schumann (flame-sheet) formulation was applied to laminar counterflow diffusion-flame experiments in which, CO was added either to the fuel stream, made up of mixtures of methane and nitrogen (N_2), or to the oxidizer stream, made up of mixtures of oxygen (O_2) and nitrogen, with both streams at normal atmospheric pressure and at normal room temperature. Experimental conditions were adjusted to fix selected values of the stoichiometric mixture fraction and the adiabatic flame temperature, and the strain rate was increased gradually until extinction occurred. At these selected sets of values, the strain rate at

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extinction was measured as a function of the CO concentration in the fuel or oxidizer stream [16]. It was found that with increasing amounts of CO in the oxidizer stream, the strain rate at extinction first increased and then decreased. With increasing amounts of CO in the fuel stream, changes in values of the strain rate at extinction was small in comparison to those measured for CO addition to the oxidizer stream. The experimental results were in agreement with predictions obtained employing detailed chemistry [16]. The main goal of the present RRA analysis is to elucidate the effect of CO addition on the structure of the reaction zone of nonpremixed methane flames, and highlight its influence on critical conditions of extinction.

The analysis described here complements a previous RRA analysis of the influence of hydrogen (H_2) on the structure of nonpremixed methane-flames [17]. It is carried out using a reduced four-step chemical kinetic mechanism that is described in Section 5. Critical conditions of extinction were predicted from results of the asymptotic analysis and were found to agree well with measurements [18]. Addition of hydrogen to methane flames was found to promote combustion by delaying extinction. An important finding of the previous asymptotic analysis was that the mechanisms by which hydrogen promotes combustion when it is added to the oxidizer stream is different from that when it is added to the fuel stream [17]. The present rate-ratio asymptotic analysis seeks to obtain a similar fundamental understanding of the influence of CO on nonpremixed methane flames. In particular the present analysis seeks to elucidate the similarities and differences in flame structures and mechanisms of extinction between CO addition to the fuel stream and CO addition to the oxidizer stream.

2. Formulation

Steady, axisymmetric, laminar flow of two counterflowing streams toward a stagnation plane is considered here. The origin of co-ordinates is placed at the stagnation point. The fuel stream is made up of CH_4 and N_2 and the oxidizer stream, is made up of O_2, and N_2. Carbon monoxide is added either to the fuel stream or to the oxidizer stream. The mass fraction of CH_4 in the fuel stream is $Y_{F,1}$, and that of O_2 in the oxidizer stream is $Y_{O_2,2}$. The temperature of the fuel stream is T_1 and the temperature of the oxidizer stream is T_2. When CO is added to the fuel stream its mass fraction is $Y_{CO,1}$ and it is $Y_{CO,2}$ when added to the oxidizer stream. Subscript 1 refers to conditions in the fuel stream and subscript 2 to conditions in the oxidizer stream. The flow-field is characterized by the strain rate, a, defined as the normal gradient of the normal component of the flow velocity on the oxidizer side of the stagnation plane [19,20]. The analysis employs a conserved scalar ξ, the mixture fraction, as the independent variable. This quantity is defined to satisfy a source-free conservation equation, $\rho(\vec{v}\cdot\vec{\nabla})\vec{v} - \vec{\nabla}\cdot[(\lambda/c_p)\vec{\nabla}\xi] = 0$, with $\xi = 0$ in the oxidizer stream far from the stagnation plane and $\xi = 1$ in the fuel stream [21–23]. Here $\vec{v}$ is the velocity vector, λ is the thermal conductivity, ρ the density, and c_p the heat capacity per unit mass of the mixture. A characteristic diffusion time, χ^{-1}, is given by

$$\chi = 2[\lambda/(\rho c_p)]|\nabla\xi|^2. \tag{1}$$

The quantity χ, termed the scalar dissipation rate [7,21], varies with ξ [24]. In the analysis the Lewis number, $Le_i = \lambda/(\rho c_p D_i)$, of the reactants, CH_4, O_2 and CO, the main products, carbon dioxide (CO_2) and water vapor (H_2O), and N_2 are presumed to be equal to unity [25]. Here D_i is the coefficient of diffusion of species i. For convenience the definitions

$$X_i \equiv Y_i W_{N_2}/W_i,$$
$$\tau \equiv c_p W_{N_2}(T - T_u)/Q_F, \tag{2}$$

are introduced. Here, Q_F is the heat released per mole of CH_4 consumed in the overall step $CH_4 + 2\ O_2 \rightarrow CO_2 + 2H_2O$, W_i is the molecular weight of species i, and $T_u = T_2 + (T_1 - T_2)\xi$. It follows that, $X_i = C_i W_{N_2}/\rho$, where C_i is the molar concentration of species i. At 298 K, $Q_F = 803{,}000$ J/mol [26].

The RRA analysis is carried out employing a reduced four step mechanism [13]. The Damköhler numbers constructed using the flow time obtained from the scalar dissipation rate, and the chemical time obtained from the rate parameters for the four-step mechanism are presumed to be large. In this asymptotic limit, chemical reactions are presumed to take place in a thin reaction zone. For complete consumption of all reactants, the reaction zone is located at $\xi = \xi_{st}$, where ξ_{st} is the stoichiometric fraction. The adiabatic temperature at ξ_{st} is represented by T_{st}. The values of ξ_{st} and T_{st} obtained from element balance and energy conservation for $Le_i = 1$ are described in Section 4.1 and are given by

$$\xi_{st} = (2X_{O_2,2} - X_{CO,2})/(2X_{O_2,2} - X_{CO,2} + X_{CO,1} + 4X_{F,1}),$$
$$\tau_{st} = X_{F,1}\xi_{st} + qX_{CO,2} + q(X_{CO,1} - X_{CO,2})\xi_{st}. \tag{3}$$

Here, $\tau_{st} = c_p W_{N_2}(T_{st} - T_{ref})/Q_F$, $q = Q_{CO}/Q_F$, and Q_{CO} is the heat released per mole of CO consumed in the overall step $CO + (1/2)\ O_2 \rightarrow CO_2$. At 298 K $Q_{CO} = 283{,}000$ J/mol [26], and $q = 0.353$. For convenience a quantity r is defined as

$$r = X_{O_2,2}(1 - \xi_{st})/(2X_{F,1}\xi_{st}) - 1. \tag{4}$$

It can be readily verified that the quantity r represents the ratio of the fraction of the flux of oxygen that consumes CO to the fraction that consumes methane. It is identical to the quantity r defined in previous studies on the influence of hydrogen on the structure and extinction of methane flames [17,18].

3. Numerical computation of flame structure

To guide the asymptotic analysis, flame structures were calculated using the San Diego Mechanism [27,28] employing a kinetic scheme made up of 470 elementary reactions among 53 species. Computations were performed with the temperatures at the boundaries $T_1 = T_2 = 298$ K, the pressure equal to 1 atm with either $Y_{CO,2} = 0$, or $Y_{CO,1} = 0$. The procedure for selecting the boundary values of the reactants, $Y_{F,1}$, $Y_{CO,1}$, $Y_{O_2,2}$, and $Y_{CO,2}$, is similar to that employed in the previous experimental and computational investigation of the influence of CO on structure and extinction of nonpremixed methane flames [16]. Eq. (3) provides two relations among the four parameters, $Y_{F,1}$, $Y_{CO,1}$, $Y_{O_2,2}$, and $Y_{CO,2}$. For CO addition to the oxidizer stream (with $Y_{CO,1} = 0$) at a selected value of $Y_{CO,2}$, the values for $Y_{F,1}$, and $Y_{O_2,2}$, were so chosen that ξ_{st} and T_{st} calculated using Eq. (3), respectively, are 0.055, and 2000 K. It follows that $Y_{N_2,2} = 1 - Y_{O_2,2} - Y_{CO,2}$, and $Y_{N_2,1} = 1 - Y_{F,1}$. A similar procedure was employed for CO addition to the fuel stream (with $Y_{CO,2} = 0$). Computations were performed using Chemkin-Pro [29]. The calculations employ mixture-average diffusion including Soret effects and heat losses by radiation.

Figures 1–3 show profiles of mass fractions of the reactants, hydrogen (H_2) and temperature, T as a function of ξ. Figure 1 shows flame structure for CO addition to the fuel stream, and Figs. 2 and 3 show flame structures for CO addition to the oxidizer stream. The cases for CO addition to the oxidizer stream have nearly the same extinction strain rates and are on either side of the peak extinction strain rate shown in Fig. 2 of Ref. [16].

The profiles in Figs. 1–3 show that CH_4 is completely consumed. It is well known that CO and H_2 are formed from consumption of CH_4 [1,3,8,13,16]. Figures 1–3 show that the mass fraction of H_2, Y_{H_2}, is small, its peak value is attained in the vicinity of the region where CH_4 is consumed, indicating that it is formed in this region, and it is primarily consumed on the left side of its peak

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