NRC Publications Archive Archives des publications du CNRC

A numerical study on the effects of CO₂/N₂/Ar addition to air on liftoff of a laminar CH₄/air diffusion flame

Guo, Hongsheng; Min, Jiesheng; Galizzi, Cedric; Escudié, Dany; Baillot, François

This publication could be one of several versions: author's original, accepted manuscript or the publisher's version. / La version de cette publication peut être l'une des suivantes : la version prépublication de l'auteur, la version acceptée du manuscrit ou la version de l'éditeur.

For the publisher's version, please access the DOI link below. / Pour consulter la version de l'éditeur, utilisez le lien DOI ci-dessous.

Publisher's version / Version de l'éditeur:

<https://doi.org/10.1080/00102202.2010.497074>

Combustion Science and Technology, 182, 11-12, pp. 1549-1563, 2010-11-01

NRC Publications Record / Notice d'Archives des publications de CNRC:

<https://nrc-publications.canada.ca/eng/view/object/?id=611e3ada-5d47-4585-b503-6c0b0e3d9a44>

<https://publications-cnrc.canada.ca/fra/voir/objet/?id=611e3ada-5d47-4585-b503-6c0b0e3d9a44>

Access and use of this website and the material on it are subject to the Terms and Conditions set forth at

<https://nrc-publications.canada.ca/eng/copyright>

READ THESE TERMS AND CONDITIONS CAREFULLY BEFORE USING THIS WEBSITE.

L'accès à ce site Web et l'utilisation de son contenu sont assujettis aux conditions présentées dans le site

<https://publications-cnrc.canada.ca/fra/droits>

LISEZ CES CONDITIONS ATTENTIVEMENT AVANT D'UTILISER CE SITE WEB.

Questions? Contact the NRC Publications Archive team at

PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca. If you wish to email the authors directly, please see the first page of the publication for their contact information.

Vous avez des questions? Nous pouvons vous aider. Pour communiquer directement avec un auteur, consultez la première page de la revue dans laquelle son article a été publié afin de trouver ses coordonnées. Si vous n'arrivez pas à les repérer, communiquez avec nous à PublicationsArchive-ArchivesPublications@nrc-cnrc.gc.ca.

National Research
Council Canada

Conseil national de
recherches Canada

Canada

This article was downloaded by: [Canada Institute for STI]

On: 1 November 2010

Access details: Access Details: [subscription number 908426518]

Publisher Taylor & Francis

Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK

Combustion Science and Technology

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713456315>

A Numerical Study on the Effects of CO₂/N₂/Ar Addition to Air on Liftoff of a Laminar CH₄/Air Diffusion Flame

Hongsheng Guo^a; Jiesheng Min^b; Cedric Galizzi^c; Dany Escudié^c; Françoise Baillot^b

^a Institute for Chemical Process and Environmental Technology, National Research Council of Canada, Ottawa, Canada ^b Laboratory CORIA, CNRS-Université et INSA de Rouen, France ^c CETHIL Centre Thermique de Lyon, CNRS-INSA-UCBL, INSA de Lyon, France

Online publication date: 27 October 2010

To cite this Article Guo, Hongsheng , Min, Jiesheng , Galizzi, Cedric , Escudié, Dany and Baillot, Françoise(2010) 'A Numerical Study on the Effects of CO₂/N₂/Ar Addition to Air on Liftoff of a Laminar CH₄/Air Diffusion Flame', Combustion Science and Technology, 182: 11, 1549 – 1563

To link to this Article: DOI: 10.1080/00102202.2010.497074

URL: <http://dx.doi.org/10.1080/00102202.2010.497074>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.informaworld.com/terms-and-conditions-of-access.pdf>

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

A NUMERICAL STUDY ON THE EFFECTS OF CO₂/N₂/Ar ADDITION TO AIR ON LIFTOFF OF A LAMINAR CH₄/AIR DIFFUSION FLAME

Hongsheng Guo,¹ Jiesheng Min,² Cedric Galizzi,³
Dany Escudié,³ and Françoise Baillot²

¹Institute for Chemical Process and Environmental Technology,
National Research Council of Canada, Ottawa, Canada

²Laboratory CORIA, CNRS-Université et INSA de Rouen, France

³CETHIL Centre Thermique de Lyon, CNRS-INSA-UCBL,
INSA de Lyon, France

The addition of exhaust gas to a combustor may cause liftoff of a diffusion flame due to several possible mechanisms. Understanding the relative influence of these mechanisms is of importance for the further development of exhaust gas recirculation combustion technology. The authors present a numerical study on the effects of CO₂, N₂ (two of primary exhaust gas components) and Ar addition on the liftoff of a laminar CH₄/air diffusion flame. A gradual switch-off approach was used to identify the relative importance of the different mechanisms. A detailed reaction scheme and complex thermal and transport properties were employed. The simulation results were validated by comparing the calculated and previously measured critical ratios of the 3 additives for liftoff.

The results show that the numerical simulation successfully reproduced the previously measured critical ratios of liftoff for all 3 studied additives. Detailed analysis of the numerical results suggests that the addition of N₂ affects flame liftoff due to the sole effect of dilution. On the other hand, the addition of CO₂ causes flame liftoff due to the dilution, thermal and chemical effects, with the dilution effect being the most significant one, followed by the thermal and chemical effects. All 3 effects tend to reduce combustion intensity and cause flame liftoff, leading to the smaller critical ratio of CO₂ than that of N₂. The radiation and transport property effects are negligible for CO₂ addition. Ar addition affects flame liftoff due to dilution, thermal, and transport property effects. However, whereas the dilution effect tends to reduce combustion intensity and cause flame liftoff, the thermal and transport property effects tend to increase combustion intensity and resist flame liftoff for Ar addition, which results in the greater critical ratio of Ar than that of N₂. Therefore, for the 3 studied additives in this paper, CO₂ has the minimum critical ratio, whereas Ar has the maximum for liftoff.

Keywords: Diffusion flame; Exhaust gas recirculation combustion; Flameless combustion; Liftoff

Received 15 September 2009; revised 29 January 2010; accepted 17 March 2010.

Address correspondence to Dr. Hongsheng Guo, ICPET, National Research Council Canada, Ottawa, Ontario, Canada K1A 0R6. E-mail: hongsheng.guo@nrc-cnrc.gc.ca

INTRODUCTION

Exhaust gas recirculation is an effective method to lower peak flame temperatures and reduce pollutant emissions. It has been widely applied in industrial applications. One technology that employs exhaust gas recirculation is high-temperature air combustion (flameless combustion), first developed by Tanaka et al. (1994). It effectively improves combustion efficiency and reduces NO_x emission by combining air preheating and exhaust gas recirculation (Guo et al., 1998; Jwüning and Jwüning, 1997). This technology has been further developed and improved in the last decade.

The introduction of exhaust gas into a combustion system may cause variations of some flame properties. One of these variations is that the addition of exhaust gas may result in the transition of a diffusion flame from attached to lifted (i.e., flame liftoff). The liftoff properties depend on the conditions of the exhaust gas injection, such as the composition and temperature of the exhaust gas. Understanding the fundamental mechanism of the effect of exhaust gas addition on flame liftoff is of great help for the further development and improvement of exhaust gas recirculation technology.

Many studies have been devoted to attached or lifted diffusion flames (Chung and Lee, 1991; Ghosal and Vervisch, 2001; Smooke et al., 1990; Takahashi et al., 2007; Takahashi et al., 1984; Xue and Ju, 2006). The effects of adding various components of exhaust gas on the formation of pollutants have also been investigated by some researchers (Bundy et al., 2003; Guo and Smallwood, 2008; Li and Williams, 1999; Liu et al., 2001; Lock et al., 2007; Park et al., 2004; Zhao et al., 2002). Generally an additive to the oxidant or fuel stream of a diffusion flame causes variations in flame liftoff and other properties due to the change in combustion intensity. The change in combustion intensity is caused by five possible mechanisms. The first one is the dilution effect that is caused by the decrease in the concentration of oxygen or fuel when an additive is in oxygen or fuel stream. The second one is the thermal effect that results from the change in specific heat of the oxidant or fuel stream due to the difference in specific heat between an additive and air–fuel. Third, some additives may participate in chemical reactions and thus modify combustion intensity. This factor is referred to as the chemical effect. In addition, some additives may alter flame temperature and combustion intensity by modifying radiation heat transfer rates in the flame. This is the fourth factor, referred to as the radiation effect. Lastly, the difference in transport properties between the exhaust gas and fuel or oxidant may also modify combustion intensity and cause variation in flame liftoff properties. This is the transport property effect.

Carbon dioxide (CO₂) and nitrogen (N₂) are two of the primary components of an exhaust gas. Most previous studies have attributed the impacts of CO₂ and N₂ addition on the extinction or liftoff of a flame to thermal and dilution effects (Lock et al., 2007; Takahashi et al., 2007; Takahashi et al., 1984). Although this is true for N₂ addition, it may not be correct for CO₂ addition. Our previous studies (Guo and Smallwood, 2008; Liu et al., 2001) showed that the addition of CO₂ affects pollutant emissions and some other flame properties due to not only thermal and dilution effects, but also a chemical effect. Takahashi et al. (2008) also noted that the addition

of CO₂ caused variations in carbon monoxide and hydrogen concentrations in the reaction kernel of laminar diffusion flames due to chemical effect. Besides, CO₂ participates in radiation heat transfer in a flame, which may also modify flame properties. Moreover, few studies have been reported on the relative importance of the thermal, dilution, chemical, radiation, and transport property effects on various properties, especially the liftoff of a diffusion flame.

The purpose of this paper is to investigate the relative importance of the five possible effects on the liftoff of a laminar methane/air diffusion flame due to the addition of carbon dioxide (CO₂) and nitrogen (N₂). The study is carried out by numerical simulation, but the results are compared with available experimental data in the literature. To help to understand the mechanism and validate the numerical model, the addition of argon (Ar) is also studied. The numerical model is described first, followed by the results of the critical ratios of additives to air at which the flame transits from attached to lifted. Then we focus on the detailed discussion of relative importance of the five possible effects of CO₂ addition. Finally, we give a short description of the mechanism of the liftoff due to the addition of N₂ and Ar.

FLAME CONFIGURATION AND NUMERICAL MODEL

Flame Configuration

The flames investigated in this paper are basically those studied experimentally by Min et al. (2008). They were generated in a combustion chamber with section area of $25 \times 25 \text{ cm}^2$ and height of 80 cm. The fuel was injected into the chamber from the center of the chamber bottom by a round stainless tube with inner and outer diameters of 0.60 cm and 1.02 cm, respectively. Oxidant was injected from the area outside the fuel tube at the bottom. The space inside this chamber is three-dimensional. However, because the section area of the chamber is much bigger than that of the round fuel tube, the wall of the combustion chamber has little effect on the flames. The formed flames were essentially two-dimensional axisymmetric flames. Therefore, in order to simplify the calculation and save computational cost, numerical simulation was carried out for two-dimensional axisymmetric flames. The simulation domain is shown in Figure 1. The size D equals 18 cm, which is smaller than the width (25 cm) of the experimental chamber, but it has been shown that further increase in D has negligible effect on simulation results. The height of the simulation domain Z_H equals 30 cm.

The base flame studied is a laminar CH₄/air diffusion flame. The inlet velocities of air and fuel in the base flame are 10 cm/s and 100 cm/s, respectively, at atmospheric pressure and room temperature (300 K) condition, being the same as those in the comparable experiment from the literature (Min et al., 2008). For other flames, the mass flow rates of air and fuel are the same as those in the base flame, whereas an additive is added to the air.

The additives studied include CO₂, N₂, and Ar. The ratio (α) of an additive in a flame is defined as the ratio of the volume flow rate of the additive to that of the air.

Figure 1 Flame configuration.

Governing Equations and Numerical Model

The numerical model solved fully elliptic governing equations for the conservation of mass, momentum, energy, and gas species mass fractions in cylindrical coordinates (r, z) . Soot was not included in the simulation because soot volume fractions in the studied flames are very small. The governing equations are the following:

Continuity:

$$\frac{\partial}{\partial r}(r\rho v) + \frac{\partial}{\partial z}(r\rho u) = 0 \quad (1)$$

Momentum:

$$\begin{aligned} \rho v \frac{\partial u}{\partial r} + \rho u \frac{\partial u}{\partial z} = & -\frac{\partial p}{\partial z} + \frac{1}{r} \frac{\partial}{\partial r} \left(r\mu \frac{\partial u}{\partial r} \right) + 2 \frac{\partial}{\partial z} \left(\mu \frac{\partial u}{\partial z} \right) - \frac{2}{3} \frac{\partial}{\partial z} \left(\frac{\mu}{r} \frac{\partial}{\partial r} (rv) \right) \\ & - \frac{2}{3} \frac{\partial}{\partial z} \left(\mu \frac{\partial u}{\partial z} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left(r\mu \frac{\partial v}{\partial z} \right) + \rho g_z \end{aligned} \quad (2)$$

$$\begin{aligned} \rho v \frac{\partial v}{\partial r} + \rho u \frac{\partial v}{\partial z} = & -\frac{\partial p}{\partial r} + \frac{\partial}{\partial z} \left(\mu \frac{\partial v}{\partial z} \right) + \frac{2}{r} \frac{\partial}{\partial r} \left(r\mu \frac{\partial v}{\partial r} \right) - \frac{21}{3r} \frac{\partial}{\partial r} \left(\mu \frac{\partial}{\partial r} (rv) \right) \\ & - \frac{21}{3r} \frac{\partial}{\partial r} \left(r\mu \frac{\partial u}{\partial z} \right) + \frac{\partial}{\partial z} \left(\mu \frac{\partial u}{\partial r} \right) - \frac{2\mu v}{r^2} + \frac{2\mu}{3r^2} \frac{\partial}{\partial r} (rv) + \frac{2\mu}{3r} \frac{\partial u}{\partial z} \end{aligned} \quad (3)$$

Energy:

$$c_p \left(\rho v \frac{\partial T}{\partial r} + \rho u \frac{\partial T}{\partial z} \right) = \frac{1}{r} \frac{\partial}{\partial r} \left(r \lambda \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left(\lambda \frac{\partial T}{\partial z} \right) - \sum_{k=1}^{KK} \left[\rho c_{pk} Y_k \left(V_{kr} \frac{\partial T}{\partial r} + V_{kz} \frac{\partial T}{\partial z} \right) \right] - \sum_{k=1}^{KK} h_k W_k \omega_k + q_r \quad (4)$$

Gas species:

$$\rho v \frac{\partial Y_k}{\partial r} + \rho u \frac{\partial Y_k}{\partial z} = -\frac{1}{r} \frac{\partial}{\partial r} (r \rho Y_k V_{kr}) - \frac{\partial}{\partial z} (\rho Y_k V_{kz}) + W_k \omega_k, \quad k = 1, 2, \dots, KK \quad (5)$$

where u and v are the velocities in axial (z) and radial (r) directions, respectively; T is the temperature; ρ is the density; W_k is the molecular weight of the k th gas species; λ is the mixture thermal conductivity; c_p is the specific heat of the mixture under constant pressure; c_{pk} is the specific heat of the k th species under constant pressure; ω_k is the mole production rate of the k th species per unit volume; and p is the pressure. Quantity h_k denotes the specific enthalpy of the k th species; g_z is the gravitational acceleration in z direction; μ is the viscosity of the mixture; Y_k is the mass fraction of the k th species; V_{kr} and V_{kz} are the diffusion velocities of the k th species in the r and z directions; and KK is the total gas phase species number.

The last term q_r on the right-hand side of Eq. (4) is the source term due to radiation heat transfer. It was obtained by the discrete-ordinates method coupled to a statistical narrow-band correlated-K (SNBCK) based band model for the properties of CO, CO₂, and H₂O (Liu et al., 1999).

The chemical reaction scheme used is GRI-Mech 3.0 (Smith et al., 1999), which is an optimized mechanism for natural gas combustion. All the thermal and transport properties were obtained by using the database of GRI-Mech 3.0 and the algorithms given by Kee et al. (1986) and Kee et al. (1980). The diffusion velocities were calculated by the mixture-average method (Kee et al., 1986). The thermal diffusion of H and H₂ was included, whereas that of other species was neglected.

Due to the symmetry, only half of the domain in Figure 1 was used in the simulations. The governing equations were discretized using the control-volume method. The low Mach number flow was assumed. The SIMPLE numerical scheme (Patankar, 1980) was used to deal with the pressure and velocity coupling. The diffusion and convective terms in the conservation equations were discretized by the central and the power law difference methods, respectively. The discretized equations of species mass fractions were solved in a fully coupled fashion on every grid to speed up the convergence process, whereas those of momentum, energy, and pressure correction were solved using the tridiagonal matrix algorithm. Variable mesh size was used, with fine mesh of $0.01 \times 0.01 \text{ cm}^2$ being put in the region around the exit of fuel tube. Outside this region, the mesh size was gradually increased by an expansion factor of 1.03.

A symmetric boundary condition was used for the center axis, and a free-slip condition was used for the outside boundary (left or right). A zero-gradient condition was used for the exit (top) of the simulation domain. For the inlet of fuel

stream, the uniform and parabolic velocity distribution profiles were tested at the beginning. However, a negligible difference was found in the results from the two inlet profiles. Therefore, all the calculation results presented in this paper were obtained by using uniform velocities and concentrations of species for the fuel-tube region and the annular oxidant stream, respectively, at the bottom of the domain.

The definition of liftoff process is discussed subsequently.

RESULTS AND DISCUSSION

Critical Ratios of Different Additives

The base flame described previously is an attached laminar diffusion flame. With the addition of CO_2 , N_2 , or Ar to air, flame temperature reduces. When the ratio (α) of an additive to air is over a critical value, flame liftoff happens. This liftoff phenomenon was investigated in numerical simulation by gradually increasing the ratio of an additive to air. The increment of the ratio of an additive is 0.01 in the calculation. If the liftoff happens between two consecutive ratios, we assume that the critical ratio (α_c) is the mean of these two numbers. For example, Figure 2 shows the mole fraction distribution of OH radical for the base flame and flames with ratios of 0.11 and 0.12 when CO_2 is added. The calculated flame at the ratio of 0.11 is attached and that at the ratio of 0.12 is lifted. As a result, we assume the critical ratio of CO_2 addition is 0.115. Therefore, the absolute error of the calculated critical ratio of an additive is ± 0.005 .

It is noted from Figure 2 that even for the base flame, the flame base is a little away from the burner rim. This is due to the cooling effect of the burner rim and the high scalar dissipation rate in the near burner rim region (Hamins et al., 1994). However, a detailed examination of numerical results shows that for the base flame and

Figure 2 Mole fraction distribution of OH radical for three flames with CO_2 addition: (a) base flame; (b) $\alpha_{\text{CO}_2} = 0.11$; (c) $\alpha_{\text{CO}_2} = 0.12$.

the flames with the ratio of an additive lower than the critical value, temperature and concentrations of radicals quickly increase with increasing distance from the burner rim along the z axis (Figure 1), whereas for the flames with the ratio of an additive greater than the critical value, the distance between the flame base and burner rim (see below) sharply increases and the temperature keeps almost constant within a certain distance above the burner rim. The distance between flame base and burner rim is referred to as the attachment height, which is defined as the vertical distance from burner rim to the location where the concentration of CH reaches 1.0% of its peak value. In this paper, we define a flame as lifted if the attachment height is more than two times the burner tube thickness (2.1 mm). Although this is an arbitrary definition, it is used for all the studied flames and therefore the results based on it should be able to reflect the relative effects of different diluents and mechanisms. Besides, due to the sharp increase once the flame is lifted, the actual attachment height for all studied lifted flames in this paper are much larger than the defined critical attachment height.

Figure 3 shows the comparison of calculated (in the present study) and measured (Min et al., 2008) normalized attachment height, which is the ratio of the attachment height from a diluted flame to that of the base flame when CO₂ is added. The measured attachment height was obtained by measuring the vertical distance between the burner rim and the location where visible CH chemiluminescence emission signal starts to appear. It is observed that the simulation successfully reproduced the observed variation trend of the attachment height. With the addition of CO₂ to air, the attachment height gradually increases until a critical ratio is reached. Then this height sharply increases and flame is lifted.

Figure 4 shows the critical ratios of CO₂, N₂, and Ar addition. The experimental data obtained by Min et al. (2008) are also shown in Figure 4. It is observed that the numerical model successfully captured the experimentally measured critical

Figure 3 Variation of normalized attachment height with increasing the ratio of CO₂ to air volume flow rates.

Figure 4 Critical ratios of CO₂, N₂, and Ar.

ratios for all three studied additives. Both experiment and numerical simulation show that the critical ratios of the three studied additives are in the order of $\alpha_{\text{CO}_2} < \alpha_{\text{N}_2} < \alpha_{\text{Ar}}$. This is qualitatively consistent with the previous observation by Takahashi et al. (2007).

As mentioned in the introduction, the addition of an additive causes flame liftoff due to five possible effects. Although understanding the relative importance of these effects is of interest for combustion scientists and engineers, little has been reported in the literature. We analyze the relative importance subsequently using the flexibility of numerical simulation.

Relative Importance of Different Effects of CO₂ Addition

For all three additives studied in this paper, their addition to a diffusion flame may cause flame liftoff due to thermal, dilution, and transport property effects. However, only the addition of CO₂ causes flame liftoff through chemical and radiation effects because N₂ and Ar are inert for chemical reactions and transparent in terms of thermal radiation. Therefore, we first give a detailed analysis on the addition of CO₂.

To identify the relative importance of these effects, five sets of simulations were carried out for CO₂ addition. In the first set of simulations (SIM1), CO₂ is gradually added to the air stream of the base flame. In the second set of simulations (SIM2), the added CO₂ is replaced by an artificial additive that has the same thermal and transport properties as CO₂ but is inert for chemical reactions. Besides, this artificial additive in SIM2 participates in radiation heat transfer in the same way as CO₂. Therefore, the effects of the additive addition in SIM2 are the same as in SIM1, except that there is no chemical effect in SIM2. The difference between the results from SIM1 and SIM2 is caused by the chemical reactions of the added CO₂ (i.e., the chemical effect). The calculation condition in the third set of simulations (SIM3) is basically the same as in SIM2, but the additive does not participate in

radiation heat transfer. However, radiation heat transfer due to other species, such as H₂O, CO, and CO₂ (combustion generated) still exists in SIM3. The difference between the results from SIM2 and SIM3 is due to the radiation effect. In the fourth set of simulations (SIM4), the specific heat of the additive is set as the same as air. Other conditions and parameters in SIM4 are the same as in SIM3. The only difference in the conditions and parameters between SIM3 and SIM4 is the specific heat of the additive. Accordingly, the difference in the results from SIM3 and SIM4 is caused by the variation in specific heat in the air stream (i.e., the thermal effect). Finally, in the fifth set (SIM5), all conditions are the same as in SIM4 except that the transport properties of the additive are the same as air. Therefore, the difference between SIM4 and SIM5 is caused by the transport property effect. Comparing SIM5 and the base flame, we note that the sole difference between them is the concentration of oxygen in the air stream since the additive in SIM5 has the same thermal and transport properties as air and does not participate in reactions and radiation heat transfer. Consequently, the base flame and SIM5 differ, owing to the dilution effect. The conditions and parameters in the previous five sets of simulations are summarized in Table 1.

Figure 5 shows the critical ratios from the five sets of simulations. The error bars are displayed based on that the increment of the ratio of the additive in the simulation is 0.01. It is observed that the critical ratio in SIM5 is 0.205. This means that the ratio of CO₂ should be above 0.205 to cause the base flame to be lifted if CO₂ addition affects the base flame only due to the dilution effect, because the additive in SIM5 does not participate in chemical reactions and radiation heat transfer, and has the same thermal and transport properties as air.

The critical ratio obtained by SIM5 is close to that for N₂ addition, which was measured by Min et al. (2008), and the same as that calculated in the present study (as shown in Figure 4). It is because the additive in SIM5 is similar to N₂. They both are inert for chemical reactions and transparent for radiation, and have similar thermal and transport properties (note that the thermal and transport properties of N₂ are very close to those of air). Therefore, the addition of both of them modifies combustion intensity of the base flame primarily due to dilution. This similarity in the critical ratios from SIM5 and N₂ addition confirms that the strategy used previously to identify the relative importance of different effects is reasonable.

Table 1 Conditions and parameters in five sets of simulations for CO₂ addition and three sets of simulations for Ar addition

	Additive participates in reactions	Additive participates in radiation	Specific heat of additive	Transport properties of additive
SIM1	Yes	Yes	Same as CO ₂	Same as CO ₂
SIM2	No	Yes	Same as CO ₂	Same as CO ₂
SIM3	No	No	Same as CO ₂	Same as CO ₂
SIM4	No	No	Same as air	Same as CO ₂
SIM5	No	No	Same as air	Same as air
ArSIM1	No	No	Same as Ar	Same as Ar
ArSIM4	No	No	Same as air	Same as Ar
ArSIM5	No	No	Same as air	Same as air

Figure 5 Critical ratios from the five sets of simulations for CO_2 addition.

The critical ratio from SIM4 is the same as that from SIM5, implying that the transport property effect on the liftoff of the base CH_4/air diffusion flame is negligible.

Compared to that in SIM4, the critical ratio in SIM3 drops to 0.135. This is because the higher specific heat of the additive in SIM3 (same as CO_2) than that in SIM4 (same as air) reduces flame temperature and combustion intensity. Therefore, the thermal effect of CO_2 addition causes the critical ratio to drop from 0.205 to 0.135. The thermal effect has been well understood and thus is not discussed in this paper.

Although the additive participates in radiation heat transfer in SIM2 but not in SIM3, the calculated critical ratios in SIM2 and SIM3 are almost same, implying that radiation effect of the added CO_2 has a negligible effect on the base flame in terms of liftoff. This is because radiation heat loss is not significant in the region near burner rim where the reaction zone is thin. However, we point out that radiation may significantly affect other flame properties in the upper flame region, such as pollutant formation.

Compared to that of 0.135 in SIM2, the critical ratio reduces to 0.115 in SIM1, meaning that the chemical effect of the added CO_2 reduces the critical ratio by about 0.02. Therefore, the addition of CO_2 affects liftoff of a diffusion flame due to not only the thermal and dilution effects, but also the chemical effect. This has never been reported in the literature. To identify what causes the chemical effect of CO_2 addition, we analyze the variation of the heat release rates in SIM1 and SIM2. The simulation indicates that the peak heat release rate in SIM1 is lower than in SIM2 (i.e., the chemical effect of CO_2 addition suppresses the heat release rate). The heat release rate is obtained by $-\sum_{k=1}^{KK} \omega_k H_k W_k$. Figure 6 displays each term inside the summation calculation for the peak heat release rate of the flame with the additive ratio of 0.10 in SIM1 and SIM2. It is found that the primary heat release is due to the formation of H_2O and CO_2 . Further, the heat release rate due to the formation of CO_2 in SIM1 is significantly lower than in SIM2, whereas that due

Figure 6 Distributions of peak heat release rate due to variations of different species in SIM1 and SIM2 for the flame with additive ratio equal to 0.10.

to the formation of H₂O in SIM1 is slightly higher than in SIM2. Therefore, the suppression of heat release due to the chemical effect of CO₂ addition is caused by the reduction in the net formation rate of CO₂ in the flame. A pathway analysis of the details of the simulation results suggests that the reduction of net CO₂ formation rate is because of the reaction $\text{OH} + \text{CO} = \text{H} + \text{CO}_2$. When CO₂ is added, the rate of the reverse reaction is intensified, resulting in a reduction in the net formation rate of CO₂ and higher concentration of CO. This is further confirmed by the variation of the maximum CO mole fraction (obtained by taking the maximum value at each section) along the height above burner exit in Figure 7, which clearly shows that the concentration of CO in SIM1 quickly becomes higher than in SIM2 once the peak value location is reached. Therefore, we can conclude that the chemical effect of the CO₂ addition on the transition of the base flame from attached to lifted is primarily caused by the reaction $\text{OH} + \text{CO} = \text{H} + \text{CO}_2$. This conclusion is qualitatively consistent with our previous studies (Guo and Smallwood, 2008; Liu et al., 2001) on the chemical effect of CO₂ addition on other flame properties. It is also not inconsistent with the finding of Takahashi et al. (2008), who indicated that the chemical effect of CO₂ addition in the reaction kernel of a diffusion flame might be due to an equilibrium in the water gas shift reactions: $\text{CO} + \text{H}_2\text{O} = \text{CO}_2 + \text{H}_2$, because $\text{OH} + \text{CO} = \text{H} + \text{CO}_2$ is the key elementary step of the water gas shift reactions.

The pathway analysis also suggests that the chemical effect of CO₂ addition on the main H₂O formation reaction $\text{H}_2 + \text{OH} = \text{H}_2\text{O} + \text{H}$ is negligible. The slightly higher formation rate of H₂O in SIM1 than in SIM2 shown in Figure 6 is due to the reactions $\text{OH} + \text{HO}_2 = \text{O}_2 + \text{H}_2\text{O}$ and $\text{CH} + \text{H}_2\text{O} = \text{H} + \text{CH}_2\text{O}$, of which the former is a formation reaction and the latter is a consumption reaction of H₂O. First, when CO₂ is added, the concentration of CH is reduced, owing to the reaction $\text{CH} + \text{CO}_2 = \text{HCO} + \text{CO}$, leading to the lower H₂O consumption rate by the reaction $\text{CH} + \text{H}_2\text{O} = \text{H} + \text{CH}_2\text{O}$. Second, because of the lower net conversion rate of CO to CO₂ in SIM1 than in SIM2, the requirement for oxygen is less and the concentration

Figure 7 Variations of the maximum CO mole fraction along the height above burner exit for the flame with ratio of additive equal to 0.10 in SIM1 and SIM2.

of oxygen in the reaction zone is slightly higher in SIM1 than in SIM2. Consequently, the formation rate of HO_2 due to the reaction $\text{H} + \text{O}_2 + \text{H}_2\text{O} = \text{HO}_2 + \text{H}_2\text{O}$ is higher in SIM1 than in SIM2. This in turn leads to the slightly higher H_2O formation rate in SIM1 due to the reaction $\text{OH} + \text{HO}_2 = \text{O}_2 + \text{H}_2\text{O}$. The higher formation rate of H_2O in SIM1 than in SIM2 can be confirmed by the higher H_2O concentration in SIM1 than SIM2 after the peak location is reached, as shown in Figure 8. Therefore, the chemical suppressive effect of CO_2 addition due to the reaction $\text{OH} + \text{CO} = \text{H} + \text{CO}_2$ is slightly moderated by the higher H_2O formation rate in SIM1 than in SIM2.

Figure 8 Variations of the maximum H_2O mole fraction along the height above burner exit for base flame and flames with ratio of diluent equal to 0.10.

Overall, we can conclude from Figure 5 that the addition of CO₂ causes the liftoff of a diffusion flame due to not only the effects of dilution and thermal, but also the modification in chemical reactions. The dilution effect is the most significant one, followed by the thermal effect. Relatively, the chemical effect is small. The radiation and transport property effects of CO₂ addition on the liftoff of a CH₄/air air diffusion flame are negligible.

Relative Importance of Different Effects of N₂ and Ar Addition

N₂ and Ar are inert for chemical reactions and transparent in terms of radiation heat transfer. Therefore, their addition to a diffusion flame only affects the flame through the thermal, dilution, and transport property effects.

N₂ has similar thermal and transport properties as does air. Therefore, its addition to the air stream only modifies the combustion intensity of a flame through the modification in the concentration of oxygen (i.e., the dilution effect). This has been confirmed by the similarity in critical ratios of N₂ calculated in the present study and measured by Min et al. (2008) to that for CO₂ addition obtained by SIM5.

In addition to the dilution effect, the addition of Ar also affects the flame due to the thermal and transport property effects. To identify the relative importance of the three effects for Ar addition, similar to CO₂ addition, three sets of simulations (ArSIM1, ArSIM4, and ArSIM5) were conducted. They correspond to SIM1, SIM4, and SIM5, respectively, for CO₂ addition. The corresponded SIM2 and SIM3 in CO₂ addition were not conducted here because Ar is inert and transparent. The conditions of ArSIM1, ArSIM4, and ArSIM5 are also summarized in Table 1. The critical ratios from the three sets are shown in Figure 9. It is observed that the dilution effect is also the most significant one. Being different from CO₂ addition, the transport property effect becomes relatively noticeable. Further, both thermal and transport property effects actually increase the critical ratio for Ar addition, based

Figure 9 Critical ratios of three sets of simulations for Ar addition.

on that due to dilution effect. The reasons are that Ar has a lower thermal conductivity and smaller specific heat than does air. The former results in lower heat loss from the reaction zone, and the latter increases flame temperature. Figure 4 shows that the critical ratio of Ar addition is significantly higher than those of N_2 and CO_2 addition. Therefore, in addition to the dilution effect, Ar addition also affects flame liftoff through thermal and transport property effects, but these two effects counter the dilution effect (i.e., they actually strengthen the capability of a flame to be attached to the burner).

CONCLUSIONS

We carried out a numerical study on the effects of three different additives to air on the transition of a laminar CH_4 /air diffusion flame from attached to lifted. We used a detailed reaction scheme and complex thermal and transport properties. The mechanisms of the transition due to the additives to air have been analyzed by a gradually switching off approach.

Three different additives were investigated and the following conclusions can be drawn.

1. The addition of N_2 affects the flame liftoff due to the sole dilution effect.
2. For CO_2 addition, it causes flame liftoff due to the dilution, thermal and chemical effects, with the dilution effect being the most significant one, followed by the thermal effect. The chemical effect is relatively small. All three effects tend to reduce combustion intensity and cause the flame to be lifted. The radiation and transport property effects are negligible.
3. For Ar, its addition to air causes flame liftoff due to the dilution effect. This dilution effect is countered by the thermal and transport property effects because of the lower thermal conductivity and specific heat of Ar than those of air.

REFERENCES

- Bundy, M., Hamins, A., and Lee, K.Y. 2003. Suppression limits of low strain rate non-premixed methane flames. *Combust. Flame*, **133**, 299.
- Chung, S.H., and Lee, B.J. 1991. On the characteristics of laminar lifted flames in a non-premixed jet. *Combust. Flame*, **86**, 62.
- Ghosal, S., and Vervisch, L. 2001. Stability diagram for lift-off and blow out of a round jet laminar jet diffusion flame. *Combust. Flame*, **123**, 646.
- Guo, H., Ju, Y., Maruta, K., Niioka, T., and Sato, J. 1998. Numerical investigations of NO_x emission in high temperature air combustion. *JSME International Journal (Series B)*, **41**, 331.
- Guo, H., and Smallwood, G.J. 2008. A numerical study on the influence of CO_2 addition on soot formation in an ethylene/air diffusion flame. *Combust. Sci. Technol.*, **180**, 1695.
- Hamins, A., Trees, D., Seshadri, K., and Chelliah, H.K. 1994. Extinction of nonpremixed flames with halogenated fire suppressants. *Combust. Flame*, **99**, 221.
- Jwnning, J.A., and Jwnning, J.G. 1997. Flameless oxidation to reduce thermal NO formation. *Prog. Energy Combust.*, **23**, 81.

- Kee, R.J., Dixon-Lewis, G., Warnatz, J., Coltrin, M.E., and Miller, J.A. 1986. A Fortran computer code package for the evaluation of gas-phase, multicomponent transport properties. Sandia Report, SAND 86-8246.
- Kee, R.J., Miller, J.A., and Jefferson, T.H. 1980. A general-purpose, problem-independent, transportable, Fortran chemical kinetics code package. Sandia Report, SAND 80-8003.
- Li, S.C., and Williams, F.A. 1999. NO_x formation in two-stage methane-air flames. *Combust. Flame*, **118**, 399.
- Liu, F., Guo, H., Smallwood, G.J., and Gülder, Ö.L. 2001. The chemical effects of carbon dioxide as an additive in an ethylene diffusion flame: Implications for soot and NO_x formation. *Combust. Flame*, **125**, 778.
- Liu, F., Smallwood, G.J., and Gülder, Ö.L. 1999. Radiation heat transfer calculations using the SNBCK method. AIAA paper 99-3679, Norfolk, VA.
- Lock, A., Briones, A.M., Aggarwal, S.K., Puri, I.K., and Hegde, U. 2007. Liftoff and extinction characteristics of fuel- and air-stream-diluted methane-air flames. *Combust. Flame*, **149**, 340.
- Min, J., Baillot, F., Domingues, E., Patte-Rouland, B., Talbaut, M., Escudié, Galizzi, C., André, F., and Gicquel, O. 2008. Effect of CO₂ addition on the stability and the flickering of non-premixed methane/air flames. 19th Int. Symposium on Transport Phenomena, Reykjavik, Iceland, 17-20 August.
- Park, J., Kim, S.C., Keel, S.I., Noh, D.S., Oh, C.B., and Chung, D. 2004. Effect of steam addition on flame structure and NO formation in H₂-O₂-N₂ diffusion flame. *Int. J. Energy Res.*, **28**, 1075.
- Patankar, S.V. 1980. *Numerical Heat Transfer and Fluid Flow*, Hemisphere, New York.
- Smith, G.P., Golden, D.M., Frenklach, M., Moriarty, N.W., Eiteneer, B., Goldenberg, M., Bowman, C.T., Hanson, R.K., Song, S., Gardiner, W.C., Lissianski, J.V.V., and Qin, Z. 1999. *GRI-Mech home page*. http://www.me.berkeley.edu/gri_mech/ (Accessed 19 August 2010).
- Smooke, M.D., Lin, P., Lam, J.K., and Long, M.B. 1990. Computational and experimental study of a laminar axisymmetric methane-air diffusion flame. *Proc. Comb. Inst.*, **23**, 575.
- Takahashi, F., Linteris, G.T., and Katta, V.R. 2007. Extinguishment mechanisms of coflow diffusion flames in a cup-burner apparatus. *Proc. Comb. Inst.*, **31**, 2721.
- Takahashi, F., Linteris, G.T., and Katta, V.R. 2008. Extinguishment of methane diffusion flames by carbon dioxide in coflow air and oxygen-enriched microgravity environments. *Combust. Flame*, **155**, 37.
- Takahashi, F., Mizomoto, M., Ikai, S., and Futaki, N. 1984. Lifting mechanism of free jet diffusion flames. *Proc. Comb. Inst.*, **20**, 295.
- Tanaka, R., Kishimoto, K., and Hasegawa, T. 1994. High temperature air combustion. *Combust. Sci. Technol.* (in Japanese), **2**, 257.
- Xue, Y., and Ju, Y. 2006. Studies of the liftoff properties of dimethyl ether jet diffusion flames. *Combust. Sci. Technol.*, **178**, 2219.
- Zhao, D., Yamashita, H., Kitagawa, K., Arai, N., and Furuhashi, T. 2002. Behavior and effect on NO_x formation of OH radical in methane-air diffusion flame with steam addition. *Combust. Flame*, **130**, 352.