Combustion and Flame 172 (2016) 13–19

Contents lists available at ScienceDirect

Combustion and Flame

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

Study on the combustion limit, near-limit extinction boundary, and flame regimes of low-Lewis-number $CH_4/O_2/CO_2$ counterflow flames under microgravity

Tomoya Okuno[a,*], Hisashi Nakamura[a], Takuya Tezuka[a], Susumu Hasegawa[a], Koichi Takase[a], Masato Katsuta[b], Masao Kikuchi[b], Kaoru Maruta[a,c]

[a] *Institute of Fluid Science, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Miyagi 980-8577, Japan*
[b] *Japanese Aerospace Exploration Agency, 2-1-1 Sengen, Tsukuba, Ibaraki 305-8505, Japan*
[c] *Far Eastern Federal University, Russky Island, Vladivostok, Russia*

ARTICLE INFO

Article history:
Received 25 January 2016
Revised 9 March 2016
Accepted 1 July 2016

Keywords:
Counterflow premixed flames
Flame ball
Radiative extinction
Flammability limit
Microgravity combustion

ABSTRACT

To obtain an accurate and comprehensive understanding on the fuel-lean combustion limit, near-limit extinction boundary and flame regimes, experimental and numerical investigations using premixed counterflow flames were conducted for low-Lewis-number mixtures. Counterflow flame experiments were conducted under microgravity ($2.2 \leqq a \leqq 12.9\ s^{-1}$) and under normal gravity ($35 \leqq a \leqq 150\ s^{-1}$) using fuel-lean $CH_4/O_2/CO_2$ mixtures ($Le = 0.75$) where a is the stretch rate. The mole fraction ratio of O_2 to CO_2 in the mixtures was 0.40. Microgravity experiments at $a = 2.7–3.2\ s^{-1}$ showed that transitions from planar flames to ball-like flames occurred near extinction for the investigated mixture. In addition to planar flames and ball-like flames, multi-dimensional flames such as cellular flames and twin-curved flames were also observed at $a = 2.2–5.5\ s^{-1}$ in microgravity experiments. In conjunction with experimental results under normal gravity, experimental flame regimes for overall stretch rates were obtained for the first time and the region where ball-like flames were observed showed a qualitative agreement with our previous study based on the 3-D transient computations with the diffusive-thermal model. Extinction points obtained by microgravity experiments were found to scatter at very low stretch rates where multi-dimensional flames such as ball-like flames and cellular flames were observed, indicating the existence of a flame regime at lower stretch rates leaner than the planar counterflow flame extinction boundary. In addition, flame bifurcation at low stretch rates and at $\phi = 0.58$ and 0.60 were experimentally observed for the present low-Lewis-number mixture, indicating the validity of the previous computational and theoretical studies on the G-shaped extinction curve of planar counterflow flames.

1. Introduction

The most important goals in combustion engineering of today are to increase efficiency of combustors and to decrease CO_2 and pollutant emission. To achieve such goals for industrial furnaces, the development of the High Temperature Oxygen combustion (HiTO*x*) furnace is under operation in Japan [1,2]. Here, CH_4 is used as the fuel and reacts with mixtures of pure oxygen and exhaust gas which contains high concentrations of CO_2 and H_2O. Combined with carbon capture and storage, it is expected to increase efficiency of furnaces and dramatically decrease CO_2 and NO*x* emission into the atmosphere. When high concentrations of CO_2 are introduced in methane flames, the Lewis number, *Le*, is much lower than unity at fuel-lean conditions and the radiation reabsorption cannot be neglected. Therefore, to develop such furnaces, an accurate and comprehensive understanding on the combustion limit, near-limit extinction boundary, and flame regimes of low-Lewis-number $CH_4/O_2/CO_2$ flames is essential.

In order to investigate the combustion limit and the near-limit boundary, the planar counterflow configuration has been widely used. For planar premixed counterflow flames with Lewis numbers lower than but close to unity, a C-shaped extinction curve has been experimentally [3] and numerically [4,5] obtained in the vicinity of the combustion limit. Steady state computations [6] and theoretical analysis [7] of planar counterflow flames at wider equivalence ratios including near-limit conditions revealed that the counterflow flame extinction boundary exhibit G-shaped curves due to flame bifurcation at low stretch rates for mixtures

---

[*] Corresponding author. Fax: +81 22 217 5296.
E-mail address: okuno@edyn.ifs.tohoku.ac.jp (T. Okuno).

http://dx.doi.org/10.1016/j.combustflame.2016.07.001
0010-2180/

with Lewis numbers lower than but close to unity. Investigations on the extinction of premixed counterflow flames, particularly addressed for $CH_4/O_2/CO_2$ mixtures have been extensively studied in the past [8–12]. However, since most of the studies have been conducted for high stretch rate conditions, the overall extinction curve including extremely low stretch rate conditions and the actual existence of bifurcation remain largely uninvestigated. This is because at low stretch rates, natural convection obscures the essential behavior of near-limit flames under normal gravity conditions. Therefore, to investigate the combustion limit and the near-limit extinction boundary for overall stretch rates, we have conducted counterflow flame experiments using $CH_4/O_2/CO_2$ mixtures under normal gravity and microgravity for stretch rates $35 \leqq a \leqq 150\,s^{-1}$ and $2.2 \leqq a \leqq 12.9\,s^{-1}$, respectively.

Using low-Lewis-number mixtures, a recent study by our group on counterflow flames under microgravity have revealed that transitions from counterflow flames to ball-like flames occur for $CH_4/O_2/Xe$ mixtures ($Le \approx 0.5$) experimentally and numerically [13]. Also, another study by our group on counterflow flames have revealed that the cellular flames are formed at low stretch rates by 3-D transient computations with one-step chemistry at $Le = 0.50$ [14]. These cellular flames are formed by diffusive-thermal instability and have also been observed using slot-jet counterflow experiments with $CH_4/O_2/CO_2$ mixtures at around $70\,s^{-1}$ under normal gravity by Liu et al. [12]. We have also shown that each cell of the cellular flames separate and close to form sporadic flames resembling flame balls at low stretch rates [14]. Flame balls were first predicted by Zel'dovich [15], and was experimentally observed under microgravity experiments using quiescent mixtures by Ronney et al. [16,17]. Kaiser et al. have also shown that cellular flames and flame tubes exist at equivalence ratios lower than the planar flame limit using slot-jet counterflow experiments with H_2/air mixtures [18], and a theoretical study on cellular flames and flame tubes by Buckmaster and Short have suggested a relationship between the single flame tube and the flame ball [19]. These studies indicate that low-Lewis-number counterflow flames are not planar and form cellular or spherical structures at low stretch rates. However, detailed investigation on the flame regimes for a wide range of stretch rates have not been conducted on $CH_4/O_2/CO_2$ counterflow flames which has a large amount of radiative reabsorption and the $Le = 0.75$.

Therefore, this paper presents the combustion limit, near-limit extinction boundary and flame regimes obtained from very low stretch rate conditions by microgravity experiments to high stretch rate condition by normal gravity experiments with $CH_4/O_2/CO_2$ mixtures where radiation reabsorption cannot be neglected. In such process, the bifurcation of counterflow flames at low stretch rate was experimentally observed.

2. Computational methods

We have conducted computations on 1-D planar counterflow flames and 1-D flame balls to identify the extinction boundaries of these ideal flames and to assess the possibility of transition from a counterflow flame to a flame ball. Computational results were also utilized to estimate the experimental conditions for microgravity experiments. $CH_4/O_2/CO_2$ mixtures were applied. The ratio of O_2 mole fraction to CO_2 mole fraction was set to be 0.40. The Lewis number for the mixtures is around 0.75. A detailed reaction mechanism, GRI-Mech 3.0 [20] without reactions related to N, were used. Ambient pressure was 1 atm.

For computation of ideal counterflow premixed flames, a PREMIX based 1-D steady state code was applied [6]. To obtain the unstable solutions, the method in [6] also uses arc-length continuations which was introduced to combustion problems by Giovangigli et al. [21]. The computational domain was 10 cm. The unburnt gas temperature at the inlet boundary was 300 K. Computations for various stretch rates at different equivalence ratios were conducted to determine the extinction boundary. Extinctions were determined to be the minimum and the maximum stretch rates on the stable solution branch.

For computations of ideal flame balls in a quiescent mixture, a PREMIX based 1-D steady state code was applied [13]. Ambient gas phase temperature was 300 K, and the computational domain was 100 cm to minimize the effect of the boundary. The justification of the boundary size can be found from [22], which states that for H_2/air flames at computation domains larger than 20 cm, only minor differences in the temperature profile were found.

For the radiation model, an optically thin radiation model (OTM) was employed for both counterflow and flame ball computations. The considered radiative species were CH_4, H_2O, CO, and CO_2. Plank mean absorption coefficients of these species were taken from [23]. In addition to the computations with the OTM, computations with the statistically narrow band model (SNB) [24] were conducted for counterflow flames to estimate the preliminary experimental conditions. Radiation from H_2O, CO, and CO_2 were considered here, and same parameters as [24] were used.

3. Experimental methods

Microgravity experiments were conducted to obtain the combustion limit, near-limit extinction boundary, and flame regimes of counterflow flames at low stretch rates. The microgravity environment was obtained by parabolic flights of an airplane (MU-300) which was operated by the Diamond Air Service Company [25], Japan. The duration of the microgravity was around 15–20 s, and the gravity levels were on the order of ±0.01 G. We have removed the data for consideration when the gravity level exceeded 0.1 G in the experimental duration since a deformation of the flame and early extinction was seen at gravity levels exceeding 0.1 G. The schematic and pictures of the experimental apparatus used in microgravity experiments are shown in Fig. 1. Two opposing burners were placed inside a cylindrical chamber with an inner diameter of 254 mm and a height of 240 mm. The counterflow burners consist of bronze circular tubes, and the burner diameter and the burner separation distance were both 30 mm. The burner diameter was chosen to be sufficiently larger than the estimated flame ball diameter.

Bronze sintered metal plates with a thickness of 3 mm and a mean mesh opening of 10 µm were installed inside the burners to obtain a laminar flow. The depth of the sintered metal plates from the burner exit ranged from 1 to 30 mm depending on the stretch rate to ensure a flat velocity profile at the nozzle exit. Detailed structure of the burner can be obtained from [3], used in microgravity experiments. $CH_4/O_2/CO_2$ mixtures were supplied to both burners forming a counterflow field. The flow rates of CH_4, O_2, and CO_2 were controlled by each mass flow controller and these three gases were mixed inside the pipes. To ensure the accuracy of the mass flow controllers, each mass flow controller is calibrated before and after each flight using a flow meter where the measurement accuracy is within ±1.0% of the reading point. This results in around ±2.0% uncertainty in the equivalence ratio and ±1.0% uncertainty in the stretch rate. Flame images were recorded with three cameras. One camera was a high-speed CCD camera (Photron MC2.1) with image intensifiers and two cameras were HD cameras. If we define a Cartesian coordinate and set the burner axis to the *x*-axis, the high speed camera was placed along the *y*-axis on the stagnation plane, and one HD camera was placed along the *z*-axis on the stagnation plane. The other HD camera was placed in a diagonal direction to the burner axis to view the whole flame. The two HD cameras were kept on record during the whole microgravity experiments. Thermocouples were fixed near the burner lip to