LEWIS NUMBER EFFECT ON EXTINCTION CHARACTERISTICS OF RADIATIVE COUNTERFLOW CH₄-O₂-N₂-He FLAMES

KAORU MARUTA,1 YIGUANG JU,2 ATSUTAKA HONDA1 AND TAKASHI NIIOKA1

1Institute of Fluid Science
Tohoku University
Kataihira, Aoba-ku
Sendai 980-8577, Japan

2Department of Aeronautics and Space Engineering
Tohoku University
Aramaki Aoba, Aoba-ku
Sendai 980-8579, Japan

This work concentrates on general extinction characteristics of low-stretched premixed flames, that is, a change in extinction curve of radiative counterflow premixed flames with the Lewis numbers. To substantiate the occurrence of the change in extinction curves based on the Lewis numbers, flammable boundaries and flame bifurcation were studied by experiment and computation with detailed chemistry.

Mixtures of methane and artificial air (O₂-N₂-He, the O₂ mole fraction being fixed at 0.21, and those of N₂ and He being varied) having four different Lewis numbers, that is, 0.97, 1.2, 1.4, and 1.8, were used. Experiments were carried out under microgravity of 10 s, and computation employing C1 chemistry was conducted taking radiative heat loss into account.

The experimental results and those of detailed computation qualitatively agreed with each other and showed a change of extinction characteristics with the increase of Lewis numbers. We call this change “G-K transition,” because the configuration of the extinction curve gradually changed from G-shaped to K-shaped with the increase of Lewis numbers. This transition clearly showed, the following; (1) the relation between counterflow flame and propagating plane flame, (2) the existence of a critical Lewis number where the lower limit of counterflow flame and the conventional 1-D limit coincide, and (3) the existence of a multiple solution region resulting from flame bifurcation.

Furthermore, two pieces of supporting evidence for flame bifurcation were obtained by microgravity experiments. One is a discontinuous extinction curve due to separated extinction curves of the K-curve for Lewis number 1.8. The other is flame pulsation due to transient flame motion in the multiple solution region initiated by the diffusive-thermal instability of high Lewis number.

Introduction

The Lewis number effect plays an important role in various aspects of combustion phenomena, such as stability of flame [1], the extinction mechanism of highly stretched flames [2-4], and so forth.

Microgravity experiments addressing the flammability limit of a gaseous fuel-lean premixture using the counterflow flame technique have revealed two kinds of flame response in a small stretch-rate region [5,6]. One is a monotonic extinction curve, and the other is a C-shaped extinction curve (a curve with a turning point) for propane/air and methane/air flames, respectively. Subsequent to the experimental observation, we investigated the C-curve numerically and explained the mechanism of radiative extinction in detail [7]. Another numerical work [8] covered two kinds of flame response. In these studies, the combined effects of radiative heat loss and Lewis numbers were discussed as a key mechanism of the phenomena. Moreover, numerical study [9] that was extended to the lower stretch-rate region yielded an overall picture of the C-curve for methane/air flame as a G-shaped extinction curve. The G-curve (shown in Fig. 2) elucidates not only the relation between the flammability limit of conventional 1-D propagating plane flame and that of stretched twin flames but also the flame bifurcation phenomena that occur in an extremely small stretch region. This complex flame response is due to the combined effects of radiative loss and Lewis number [9]. A theoretical work [10] addressing the experimental results for methane/air flame also covered this region and found an “ilet” in addition to the “isola” response. Recently, we conducted a numerical calculation [11] employing a one-step overall reaction to clarify changes of extinction characteristics with general Lewis numbers systematically. We found that a G-shaped curve was gradually deformed and changed into a K-shaped curve (shown in Fig. 5)
with the increase of Lewis numbers, which we termed the G–K transition. Examination of these curves [12] indicated the possibility of multiple flame speeds in turbulent combustion.

In the present stage, however, G–K transition is not covered by other means such as detailed computation or experiments. Also, the flame bifurcation phenomenon itself has not yet been fully realized by experiments.

In the present study, both microgravity experiments and numerical calculation with detailed chemistry were carried out to examine G–K transition. We have also attempted to present supporting evidence for flame bifurcation based on experiments. By this, much more information on the role of Lewis number in radiative counterflow flame is expected.

Microgravity Experiments

The experimental apparatus has been described in our previous work [6] and hence only an outline is given here. The system consists of a counterflow pipe burner, 20 mm in diameter, mass flow controllers, a notebook PC, D/A and A/D converters, a thermocouple for recording gas temperature at the burner exit, air and fuel containers, and a video system, as shown in Fig. 1. Four compositions of artificial air consisting of O2-N2-He were used to elucidate the effect of Lewis numbers. The oxygen concentration of all these airs was fixed at 0.21, and helium/nitrogen concentrations were 0/0.79, 0.11/0.68, 0.21/0.58, and 0.46/0.33. Hence, Lewis numbers of the fuel-lean mixtures of these airs with methane were 0.97, 1.2, 1.4, and 1.8, respectively. These airs were prepared by the partial pressure method and bottled in containers in advance. To obtain an extinction within 10 s, the fuel gas concentration of the mixture supplied to the burner was gradually decreased until extinction, maintaining a constant total flow rate. In this study, discussion based on the stagnation-point velocity gradient defined by \( \frac{2U}{L} \) is employed for purposes of simplicity [6], where \( U \) and \( L \) are the flow velocity of the mixture at the burner exit and the spacing of the two pipes, respectively. Fuel concentration at extinction is considered to be the extinction limit at that stagnation-point velocity gradient, and the extinction limit curve can thus be plotted on the (stagnation-point) velocity gradient–equivalence ratio plane. Experiments on (stagnation-point) velocity gradient smaller than 25 s\(^{-1}\) were performed under microgravity at the JAMIC in Hokkaido, Japan. An apparatus experiences microgravity of 10 s during a free-fall in the 490-m drop shaft.

Computation

The mathematical model and numerical code that were employed are the same as those we previously used [7,9], based on the work by Giovangigli and Smooke [13]. An optically thin model was employed to consider the radiative heat loss from CO\(_2\) and
EXTINCTION CHARACTERISTICS OF COUNTERFLOW CH$_4$-O$_2$-N$_2$-He FLAMES

Results and Discussion

To investigate the effect of Lewis numbers on extinction characteristics, we prepared four kinds of artificial air by changing their helium gas dilution ratio. By doing this, mixture strength might be altered with the Lewis number, resulting in nonconstant adiabatic flame temperatures and conventional flammability limits. However, the effect of Lewis number on extinction characteristics themselves, rather than the value of conventional flammability limit or the velocity gradient at the limit, is the focus of the following discussion.

Extinction Curve for $Le = 0.97$

It is indispensable to employ a temperature response as a function of velocity gradient when the opening up of a solution, extinction, and jump limit for each Lewis number are fully discussed. However, to emphasize that the change of extinction limits depends on Lewis numbers, only extinction curves on the velocity gradient–equivalence ratio plane will be given here. The relation between temperature response and extinction limits were previously discussed in detail [9,11].

Figure 2 shows extinction limits as a function of equivalence ratio when the Lewis number is 0.97, that is, a mixture of methane and normal air. Although this result was previously presented [9,11], we indicate it again for purposes of comparison. Data obtained by microgravity experiments and the results of numerical calculation are plotted in the same figure. In the case of $Le = 0.97$, the results obtained by the simple one-step reaction model agreed well with these results as already confirmed elsewhere [11]. Upstream heat loss was confirmed to be negligible, based on measurement of temperature at the burner port.

The extinction curve obtained was not monotonic but exhibited a complex configuration owing to the combined effects of radiative heat loss and Lewis number. We named it the G-shaped curve [9,11]. The flammable region is the area on the right-hand side of the curve ABCDE in this figure [9,11]. There is another stable solution of flame, called weak flame, within the area bounded by the curves GD, DE, and BCF. Thus, two kinds of stable solutions are possible in this area [9,11]. Furthermore, the lower limit of this mixture is not given at point E but at turning point B because the conventional flammability limit of the 1-D propagating plane flame E is much richer than point B [9,11]. Although quantitative agreement between experimental and computational findings was not perfect, the tendency of the extinction curve, that is, the experimental C-curve, supported the computational results qualitatively [9]. Also, the turning point of the experimental C-shaped curve is leaner than the flammability limit of the 1-D propagating plane flame equal to $\phi = 0.51$ based on microgravity experiments [17].

Extinction Curve for $Le = 1.2$ and 1.4

Figures 3 and 4 show extinction limits as a function of equivalence ratios when Lewis numbers are 1.2 and 1.4, respectively. As with the case of $Le = 0.97$, agreement between experimental and computational findings is fair, suggesting the validity of H$_2$O. The CHEMKIN code [14–16] and the C$_1$ elementary reaction mechanism, which involves 58 reactions and 18 species, given by Kee et al. [16], were used.
Fig. 4. Extinction limits as a function of equivalence ratio when \( Le = 1.4 \): computational results (solid curves); experimental data obtained in normal gravity (○) and microgravity (●).

Fig. 5. Extinction limits as a function of equivalence ratio when \( Le = 1.8 \): computational results (solid curves); experimental data obtained in normal gravity (○) and microgravity (●).

Both of them. Due to the experimental procedure we employed, extinction limits corresponding only to the computational curve ABC were observed experimentally.

Figures 3 and 4 both have a turning point and weak flame solutions within the small velocity gradient region. However, flame strengthening due to the Lewis number effect weakened with the increase of Lewis numbers. Thus, it should be noted that the equivalence ratio at the flammability limit of the 1-D propagating plane flame E is greater than that at turning point B in the case of \( Le = 1.2 \), as shown in Fig. 3. The former becomes less than the latter, however, in the case of \( Le = 1.4 \), as shown by Fig. 4. This means that the lower limit switches from the equivalence ratio at turning point B to that at 1-D limit E depending on its Lewis number. These figures suggest that the critical Lewis number for this switching is a little less than 1.4. Although the general tendency of the two extinction curves obtained by the simple reaction model [11] agreed with those of Figs. 3 and 4, the critical Lewis number based on the simple reaction model is about 1.25. The critical Lewis number should be examined carefully when the flammability limit is discussed in relation to fire safety.

Extinction Curve for \( Le = 1.8 \)

Figure 5 shows extinction limits as a function of equivalence ratio when the Lewis number is 1.8. It is noted that the shape of this curve is different from the case of other Lewis numbers. First, with regard to curve ABE, the stretch limit of normal flame is separated from the weak flame branch and exhibits a monotonous extinction curve. Second, the equivalence ratio at the 1-D limit E is the lower limit of this mixture in this case. This mixture has the same tendency as a propane–air mixture [5] whose Lewis number is also around 1.8. Agreement between experimental and computational findings is fair at this as well as at the other Lewis numbers, but it should be noted that the experimental extinction curve is discontinuous, unlike the case of the other Lewis numbers. From the comparison between experiments and calculations, the authors suppose that experimental data for velocity gradients greater than 10 s\(^{-1}\) belong to the curve AB, which is the stretch limit of normal flame, and those of velocity gradients less than 10 s\(^{-1}\) belong to the curve DHIC, which is the limit of weak flame. If this is true, the reason why this discontinuous curve was observed experimentally is again the procedure we employed. Namely, the fuel concentration of the mixture decreases until extinction and thus the velocity gradient remains constant. Consequently, this discontinuous curve can be regarded as experimental evidence of flame bifurcation. Other than the existence of a multiple solution region, extinction induced by the diffusive-thermal instability of high Lewis number [18] might be related to this result. In other words, extinction due to instability might occur before flame reaches its extinction limit of the solution branch. However, because discontinuity is very large, the existence of a multiple solution region seems to be an appropriate interpretation in this case. We expect that this is the first experimental observation of the flame bifurcation of weakly stretched flames. In the next section, other phenomena related to the flame bifurcation will be presented.

Flame Behavior near Extinction

Figure 6 shows representative flame behavior during microgravity extinction experiments. Variations
of distance between twin flames with time were plotted. As shown in Fig. 6, the distance between twin flames becomes smaller with a decrease in the equivalence ratio. In the case of a Lewis number equal to 0.97, the gradient of distance between twin flames is constant until extinction (type 1). When the Lewis number is 1.8, however, the gradient of distance between the twin flames suddenly increases before extinction (type 2). Because of the well-known existence of two kinds of extinction modes of counterflow premixed twin flames [2–4], type 1 seems natural, but type 2 does not, because the twin flames of \( \text{Le} = 1.8 \) do not vanish until they merge with each other. Our interpretation of type 2 is as follows. Taking extinction induced by the diffusivethermal instability [18] into account, in this case, the flame moves along the stable branch of the steady-state solution and reaches a critical point (point A in Fig. 7) where the steady-state solution becomes unstable. Also, the distance between twin flames is not zero in this case. Hence, flame may change from the steady state and enter a flame in transient state because it is within the flammable region. Thus, the flame on the right-hand side of point * in Fig. 6b appears to be a transient flame. The extinction that occurred at * in Fig. 6b corresponds to instability-induced quenching [18]. In the case of \( \text{Le} = 0.97 \) and 1.2, type-1 extinctions were observed, and in the case of \( \text{Le} = 1.8 \), type-2 modes were observed. Our observation disagrees with that of the stretch-induced extinction of high Lewis number flame, in which the flame vanishes immediately at a certain distance. The difference in time scale, however, may be one possible cause of the disagreement.

Based on the above-mentioned assumption, the extinction curve obtained by the experiment, for instance, that shown in Fig. 5, was plotted by considering point * in Fig. 6b as an extinction limit, which may be equivalent to the instability-induced extinction limit [18]. So strictly speaking, this extinction limit is different from point B in Fig. 7, which was used as an extinction limit when the computational results were plotted. Consequently, the computational and experimental extinction curves in these figures should not be directly compared when the extinction mode is type 2. In fact, point * in type 2 is not an extinction point but rather the transition from the stable flame of the steady-state solution to the transient flame. Stability analysis for radiative counterflow flames is imperative.

Although the foregoing discussion becomes very important when flame pulsation is discussed in the next section, it does not affect the result of the discussion for Figs. 2–5 concerning the change of extinction curves.

**Observation of Flame Pulsation**

Twin flames were stable during microgravity experiments, and the flame movement perfectly obeyed control of fuel concentration [6] with a notable exception. That is, we found flame pulsation phenomena when \( \text{Le} = 1.4 \) in a particular region of velocity gradient and equivalence ratio. In this section, we introduce flame pulsations observed by the experiments and try to find a plausible explanation for them.

Figure 8 shows the variation of distance between twin flames when the Lewis number is 1.4. The mean flow velocity at the burner exit and the equivalence ratio of the mixture are also indicated in the same figure. Different from the above-presented extinction experiment, both mean flow rate and fuel
concentration were kept constant after 5.5 s from the beginning of the drop. Although the condition of the mixture was kept constant, twin flames pulsed periodically as shown in Fig. 8. Flames remained flat during the pulsation, and only the distance between the twin flames oscillated. Here, note that this pulsation begins after the type-2 behavior indicated by point * in Fig. 8. That is, after transition to transient flame, the mixture condition remained constant. The time delay for mixture transportation from the mass flow controllers to the flame has been accounted for and already compensated in Fig. 8 as was done in our previous study [6].

Figure 9 denotes the points where flame pulsation occurs. In the present experiment, it was observed only at the indicated points in Fig. 9. Reproducibility of flame pulsation at these points was confirmed by repeating the experiments. In Fig. 9, these points seem to be outside of the weak flame region shown in Fig. 4. However, it should be strongly emphasized that they are not outside but rather inside the multiple solution region. That is, they are above the curve BCF of Fig. 4 because of the type-2 behavior discussed in the previous section. The present computation indicated that four solutions, including unstable branches, are possible where these points exist.

Because each stable flame solution corresponds to each stable distance between twin flames, the pulsation might be considered as transient flame motion. To investigate this transient flame motion further, the distance between twin flames, \( x \), which represents flame solution as a state variable and its time derivative, \( \dot{x} \), are plotted as shown in Fig. 10. It should be noted that this figure is a projection of the phase trajectory onto the \( x-\dot{x} \) plane showing the transient behavior of twin flames. From this figure, the existence of two equilibrium states are indicated as well as the transient motion of flame around these two states. Unfortunately, however, it is impossible to tell whether they are stable or unstable only from this figure.

From Figs. 8–10, we now try to construct a plausible interpretation of the flame pulsation. The triggering process seems to be a transition from the steady-state stable flame to transient flame due to the diffusive-thermal instability of high Lewis number. That is, with the decrease of equivalence ratio before 5.5 s in Fig. 8, the flame becomes unstable.
at point * in Fig. 8 and departs from the state of equilibrium as with type 2 in Fig. 6b. After that, by accident in this case, other solutions are available in the smaller region of the distance between the twin flames. Namely, the flame moves around the two solutions just like the phase trajectory of the typical system with two equilibrium states. Because microgravity lasts only 10 s, we have no way to investigate whether these equilibrium states are stable or unstable experimentally. In the present stage, therefore, we cannot identify which solution branch corresponds to the two equilibrium states in Fig. 10.

Relevant unstable premixed flame phenomena were also observed by Pearlman and Ronney [19]. However, to the authors’ knowledge, the present experiment is possibly the first observation of flame moving around two equilibrium states.

To examine these lengthy scenarios, we need stability analysis, investigation of transient flame, and a longer duration of microgravity. If the scenario is true, the present result is possibly the first experimental evidence showing that flame can shift to a state of chaos close to the extinction, information necessitating changes in turbulent flame modeling.

Conclusions

Microgravity experiments and computation employing detailed chemistry were carried out to examine G–K transition, i.e., a change in extinction characteristics of radiative counterflow premixed flames with the increase of Lewis numbers. Using mixtures of methane and artificial air composed of O₂, N₂, and He, four Lewis numbers were chosen for the investigation. The results of the present computation qualitatively agreed with those of microgravity experiments and covered a wider range of velocity gradient and equivalence ratio than experiments. As a result, it was substantiated that a G–K transition, i.e., a change in extinction characteristics of radiative counterflow premixed flames with the increase of Lewis numbers. Namely, the flame moves around the two solutions just like the phase trajectory of the typical system with two equilibrium states. Because microgravity lasts only 10 s, we have no way to investigate whether these equilibrium states are stable or unstable experimentally. In the present stage, therefore, we cannot identify which solution branch corresponds to the two equilibrium states in Fig. 10.

Relevant unstable premixed flame phenomena were also observed by Pearlman and Ronney [19]. However, to the authors’ knowledge, the present experiment is possibly the first observation of flame moving around two equilibrium states.

To examine these lengthy scenarios, we need stability analysis, investigation of transient flame, and a longer duration of microgravity. If the scenario is true, the present result is possibly the first experimental evidence showing that flame can shift to a state of chaos close to the extinction, information necessitating changes in turbulent flame modeling.

Acknowledgments

The authors would like to thank Prof. Hideaki Kobayashi and Prof. Toshiyuki Hayase of Tohoku University for their stimulating discussion on transient flame phenomena and Mr. Susumu Hasegawa and Mr. Katsuoshi Muso for their assistance with the microgravity experiments. This work was performed under the management of the Japan Space Utilization Promotion Center as a part of an R&D project of Advanced Furnaces and Boilers supported by the New Energy and Industrial Technology Development Organization.

REFERENCES