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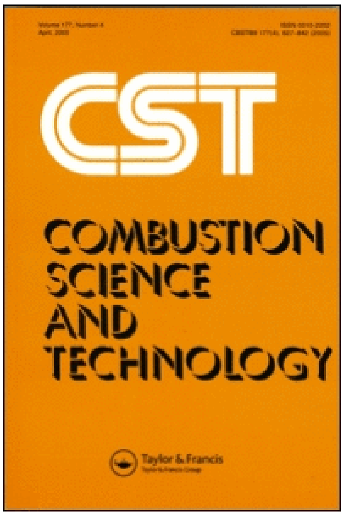

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Numerical Investigation of CH_4/CO_2/Air and $CH_4/CO_2/O_2$ Counterflow Premixed Flames with Radiation Reabsorption

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Effects of radiative heat loss on temperatures and extinction characteristics of CH_4/CO_2/air and $CH_4/CO_2/O_2$ counterflow premixed flames were numerically investigated by using the detailed chemistry and transport properties with emphasis on assessing the importance of radiation reabsorption. Radiative transfer was calculated using the discrete ordinate method along with the grey gas assumption. Results show that radiation reabsorption has little influence on the temperatures and extinction limits of CH_4/ air counterflow premixed flames of low equivalence ratio. However the radiation reabsorption has a significant effect on the flame temperatures and extinction limits of CH_4/CO_2/air and $CH_4/CO_2/O_2$ counterflow premixed flames, especially on the radiation extinction limits. The flammable region of CH_4/CO_2/air counterflow premixed flames becomes smaller as the fraction of CO_2 added to air increases. Radiation extinction of $CH_4/CO_2/O_2$ flames occurs at higher stretch rates than that of CH_4/air flames.

Keywords: Laminar premixed flames; radiation; extinction

INTRODUCTION

Flammability limits and the associated limiting mechanisms of premixed flame are of great interest to combustion scientists. For an one-dimensional premixed flame in a doubly-infinite domain, it has been well known that

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radiative heat loss produces a concentration limit (Spalding, 1957; Lakshmisha, 1990). For stretched premixed flames, experimental and theoretical investigations showed that an excessive stretch can cause a flame to extinguish due to the incomplete combustion (Tsuji and Yamaoka, 1982; Sato, 1982). However, the effect of radiation heat loss on stretched flames has not been adequately known. On the other hand, the development and application of laminar flamelet concept to turbulent flame simulation require a detailed understanding of the structure and extinction mechanisms of stretched flames. It is therefore of importance to examine the effects of radiative heat loss on stretched flames.

Sohrab and Law (1984) investigated the effect of radiative heat loss on stretched premixed flames, and concluded that radiation has little impact on the extinction of stretched flames because the stretch rate in their study was not extended to a sufficiently low value. Egolfopoulos (1994) also studied the radiation effect on steady and unsteady stretched flames, and indicated that radiation is important for near limit stretched laminar flames and for weakly stretched diffusion flames. However, the study was not extended to sufficiently low stretch rates either. Platt and T'ien (1990) first investigated the extinction of counterflow premixed flames at low stretch rates, and found that the flame extinguished at a low stretch rate because of the radiation heat loss. However, a simple one-step overall reaction mechanism, which cannot consider the effects of chain-branching and chain-terminating process, and constant transport properties were used in the paper. The experimental study carried out by Maruta *et al.* (1996) under microgravity conditions indicated that, in addition to the stretch extinction which occurred at a high stretch rate, the flame extinction also occurred when the stretch rate decreased to a sufficiently low value. Numerical investigations by Guo *et al.* (1997) using the detailed chemistry and transport properties indicated that the flame extinction at a low stretch rate is due to radiation heat loss and obtained a C-shaped extinction limit curve. More recently, Ju *et al.* (1997) found the phenomenon of two stable flame branches and obtained a G-shaped curve in their numerical study. The existence of two stable flame branches was also shown theoretically by Buckmaster (1997) based on a simple asymptotic analysis. Nevertheless the radiation reabsorption was ignored in all above investigations. Since the flame thickens and the optical thickness of flame increases as the stretch rate decreases, there have been concerns on whether or not this extinction limit induced by radiation heat loss still exists when the radiation reabsorption is taken into account and on how good the optically thin assumption is in the study of counterflow premixed flames. Therefore it is necessary to further study the radiation extinction phenomenon by using a better radiation model in which the radiation reabsorption is taken into account.

Furthermore, the flue gas recirculation, which can suppress NO_x emission and improve heat transfer in combustion chambers, is an important technique in industrial applications. CO_2 is the main species in the flue gas, and is also a main absorbing and emitting species in flames. Ronney and his coworkers did excellent works (Abbud Madrid and Ronney, 1990; Ronney *et al.*, 1994) on the addition of CO_2 to a combustible mixtures by flame ball study. However no detailed examination of the effect of CO_2 addition on stretched premixed flames has been conducted. It is of great interest to examine the effects of adding CO_2 to the gas mixture on both the stretch and radiation extinction limits of counterflow premixed flames. Two scenarios were considered for this purpose: (1) adding CO_2 to air, and (2) replacing N_2 in air with CO_2.

The objectives of the present numerical investigation are twofold. First, the questions concerning the validity of the optically thin assumption in the calculations of CH_4/air counterflow premixed flames are answered. The emphasis is on the region of low equivalence ratio. Secondly, the effects of CO_2 addition in the two scenarios mentioned earlier are investigated.

NUMERICAL MODEL

The simulation for counterflow premixed flames assumed a laminar stagnation point flow. Twin flames are formed near the stagnation plane of two opposed-jet flows, as shown in Figure 1. The cylinder coordinate system is used. Half the domain is solved due to the symmetry of this flame configuration. By assuming the stagnation point flow approximation (Giovangigli and Smooke, 1987; Kee *et al.*, 1988), the governing equations can be written as

$$\begin{aligned}
\frac{da}{dt} &= 0, \\
\frac{d\rho}{dt} + \frac{dV}{dy} &= -2\rho G, \\
L(G) &= \frac{d}{dy}\left(\mu\frac{dG}{dy}\right) - \rho G^2 + \rho\left(\frac{da}{dt} + a^2\right), \\
C_p L(T) &= \frac{d}{dy}\left(\lambda\frac{dT}{dy}\right) - \sum_{k=1}^{KK} \rho Y_k V_k C_{pk}\frac{dT}{dy} - \sum_{k=1}^{KK} h_k \omega_k M_k + q_r, \\
L(Y_k) &= \frac{d}{-dy}\left(\rho Y_k V_{ky}\right) + \omega_k M_k
\end{aligned} \tag{1}$$

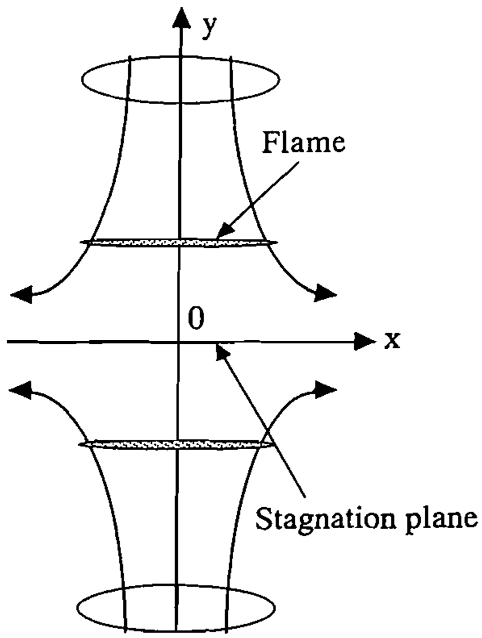

FIGURE 1 Schematic graph of the computational model.

where $L(\phi) = d\phi/dt + Vd\phi/dy$, t is the time, y and x denote the axial and radial coordinates, V is the axial mass flow rate and a is the stretch rate. G is a combined function of the stretch rate and the stream function; ρ, T and Y_k are respectively the mass density, temperature and mass fraction of kth species; μ, C_{pk} and M_k denote the mixture viscosity, the constant pressure heat capacity and the molecular weight of the kth species; h_k, V_{ky} and ω_k are respectively the specific enthalpy, the diffusion velocity in y direction and the molar production rate of the kth species. The quantity q_r is the sink term due to thermal radiation, for which the calculation method is different from that in our previous investigations (Guo et al., 1997; Ju et al., 1997) and will be described later, and KK is the number of species.

The potential boundary conditions were used. They are given as

$$y = -L; \quad T = T_L, \ Y_k = Y_{kL}, \ G = a$$
$$y = 0; \quad dT/dy = 0, \ dY_k/dy = 0, \ dG/dy = 0, \ V = 0$$

where the subscript L represents the data at the burner exit.

The C1 elementary reaction mechanism, which involves 58 reactions and 18 species, given by Kee *et al.* (1994) was employed. Our calculations were carried out basically by using the code developed by Smooke *et al.* (Smooke, 1982; Giovangigli and Smooke, 1989; Kee *et al.*, 1994). Windward difference was used for the convective term, and adaptive refinement of meshes was done. An improved arc-length continuation method (Giovangigli and Smooke, 1989; Kee *et al.*, 1988; Ju *et al.*, 1997) was used to obtain the extinction limits. The distance between burners was kept as 50 cm in all the calculations. The pressure and environment temperature were 1 atm and 300 K, respectively.

Assuming the gas mixture is grey, q_r is calculated as (Modest, 1993):

$$q_r = -k_p\left(4\sigma T^4 - \int_{4\pi} I d\Omega\right) \quad (2)$$

where k_p is the Planck mean absorption coefficient, I is the radiation intensity, Ω is the solid angle and σ is the Stefan–Boltzmann constant. The second term on the right hand side of Eq. (2) represents the contribution of thermal radiation from the entire surroundings. This term is neglected when the optically thin assumption is made and the environment temperature is cold. The contribution of this integral term is named "reabsorption" in this paper.

The Planck mean absorption coefficient was used in the calculations for the reason that results of the present investigation can be compared directly with those of our previous study (Guo *et al.*, 1997) to evaluate the importance of radiation reabsorption term when the same absorption coefficient is used. The four important radiating gaseous species in CH_4/air flame are CO_2, H_2O, CO and CH_4, and were taken into account in the evaluation of Planck mean absorption coefficient of the mixture, *i.e.*,

$$k_p = p\sum x_i k_{p,i} \quad (3)$$

where p is pressure (atm), $k_{p,i}$ and x_i denote, respectively, the mean absorption coefficient and mole fraction of absorbing and emitting species. The quantity $k_{p,i}$ was obtained by fitting the data given by Tien (1967).

In order to account for the radiation reabsorption term in the present study, the entire radiation intensity field has to be calculated. This is done by solving the following radiative transfer equation

$$\mu\frac{\partial I}{\partial y} = -k_p I + k_p I_{b,g} \quad (4)$$

using the discrete ordinates method. Where μ is the direction cosine, and $I_{b,g}$ is the black body radiation intensity at the local medium temperature. The spatial and angular discretizations of Eq. (4) was achieved by using the diamond and the S_6 schemes. Further details of the discrete ordinates method can be found in Fiveland's work (1984).

RESULTS AND DISCUSSIONS

Unless it is otherwise indicated, all the calculations were conducted by using the present radiation model, which can consider the radiation reabsorption. The results of optically thin model were taken directly from our previous study (Guo *et al.*, 1997).

CH_4/Air Flames

The temperature variations of two equivalence ratio flames *versus* the stretch rate calculated with and without radiation reabsorption are shown in Figure 2. It can be found that there are two extinction limits for every flame: the radiation extinction limit induced by radiation heat loss and the stretch extinction limit induced by excessive stretch. The existence of these two extinction limits has been discussed in our previous studies (Guo *et al.*, 1997; Ju *et al.*, 1997)

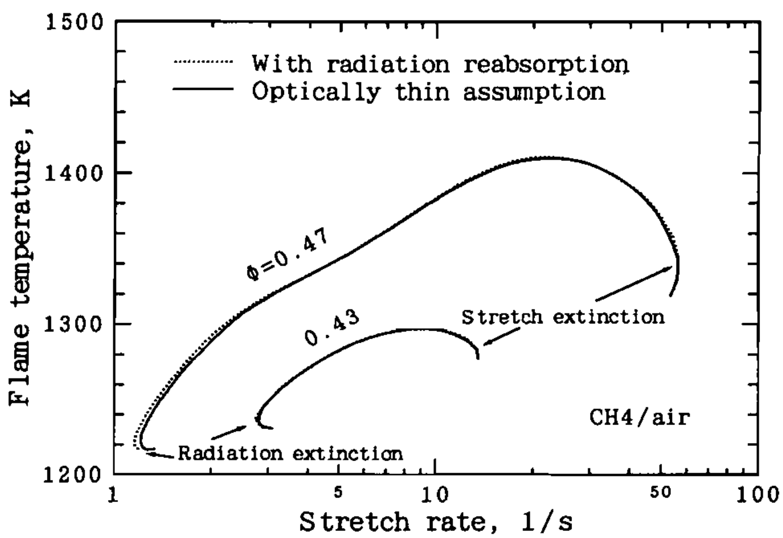

FIGURE 2 Comparisons of flame temperatures at equivalence ratios 0.43 and 0.47.

along with the mechanisms of radiation extinction. These results indicate that the radiation extinction limit still exists when the radiation reabsorption is taken into account.

We can note, from Figure 2, that the reabsorption has almost no effect on the flame temperature for the equivalence ratio of 0.43. There is only a slight influence of the radiation reabsorption when the stretch rate is decreased to near the radiation extinction limit for the equivalence ratio of 0.47. This observation can be explained as follows. For lower equivalence ratio flames, the optical thickness of flame within all flammable stretch rate range is very small, and thus the reabsorption of radiation is negligible and the optically thin assumption is appropriate. For larger equivalence ratio flames, the flame thickness and the optical thickness of flame relatively increase, especially when the stretch rate decreases to a sufficiently low value. Therefore the reabsorption begins to affect the flame temperature. However, even for the equivalence ratio of 0.47, the reabsorption has only a slight influence when the stretch rate is decreased to near the radiation extinction limit, as shown in Figure 2.

The C-shaped curve (Guo *et al.*, 1997), is an extinction limit curve indicating the flammable region of counterflow premixed flames. The calculated C-shaped curves with and without radiation reabsorption are compared in Figure 3. It can be found that the radiation reabsorption has

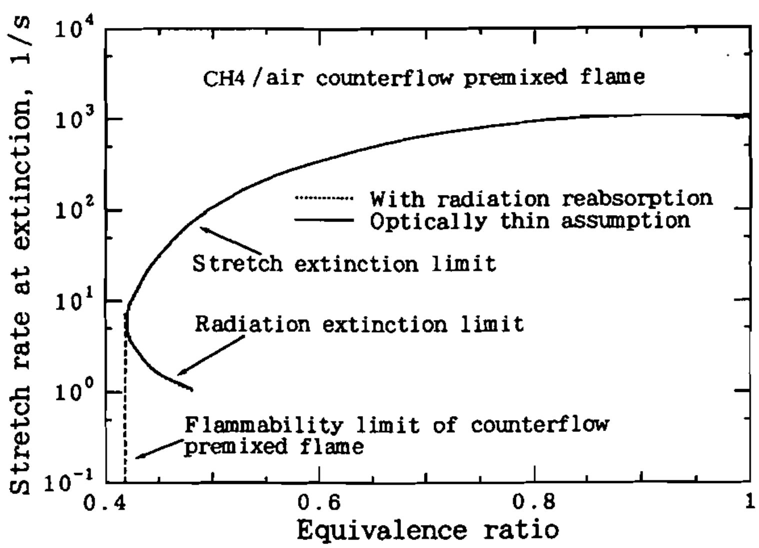

FIGURE 3 The C-shaped curve of CH_4/air flame.

almost no effect on the branch of stretch extinction limit and has little effect on the branch of radiation extinction limit when the equivalence ratio is less than 0.49. As we have indicated in our previous investigations (Guo *et al.*, 1997; Ju *et al.*, 1997), the radiation extinction limit does not exist when the equivalence ratio is greater than 0.49. Therefore we can conclude that the radiation reabsorption has little effect on the radiation extinction limit, and thus has little effect on the C-shaped curve of CH_4/air counterflow premixed flames.

CH_4/CO_2/Air Flames

Since CO_2 is an emitting and absorbing species, the optical thickness of flame will increase if CO_2 is added to the mixture. Figure 4 is the comparison of flame temperatures between the case with the radiation reabsorption and the case without the radiation reabsorption, where the fraction of CO_2 added to air is 10% and the concentration of CH_4 in the mixture is 4.8%. It is evident that the reabsorption has a significant influence on this kind of flame, and this influence becomes more pronounced as the stretch rate decreases. Therefore the reabsorption must be taken into account in the calculation of CH_4/CO_2/air counterflow premixed flames, *i.e.*, the optically thin assumption is no longer valid.

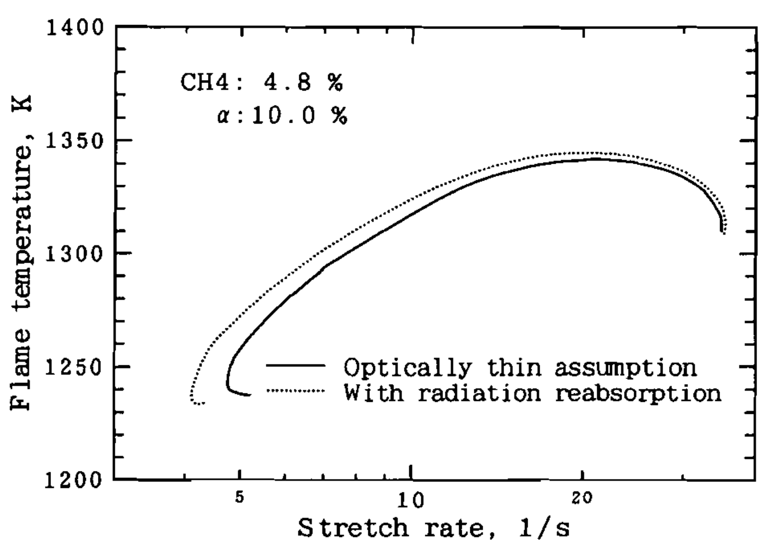

FIGURE 4 The comparison of flame temperature, where α is the fraction of CO_2 added into air.

By using the present radiation model, we extended our investigations into CH_4/CO_2/air counterflow premixed flames for the fraction of CO_2 added to air being 0.0, 5.0 and 10.0% respectively. Figure 5 gives the temperatures of flames for the fraction of CO_2 added to air being 0.0, 5.0 and 10.0%, respectively, while keeping the CH_4 percentage in the mixture constant 4.7. It can be seen that the flame temperature drastically decreases and thus the flammable stretch rate range narrows as the CO_2 fraction increases, regardless of the fact that the equivalence ratio actually increases (being 0.47, 0.49 and 0.52 respectively). It should be noted that the reduction of stretch rate at the stretch extinction limit, on which the radiation heat loss has little effect, is much more significant than the increase of stretch rate at the radiation extinction limit induced by the radiation heat loss. This phenomenon is not contributed by the radiation heat loss but by the increase of heat capacity of the mixture when the CO_2 fraction is increased. Since CO_2 has a larger heat capacity than air, the mixture heat capacity increases as the fraction of CO_2 added to air is increased. As a consequence, the flame temperature reduces as the fraction of CO_2 added to air increases. Therefore the flame is more prone to extinguish at both the stretch extinction limit and radiation extinction limit. Meanwhile, the flame is very thin at a higher stretch rate, so the residence time is very short near the stretch extinction limit. Since the flame thickens as the stretch rate decreases, the residence time

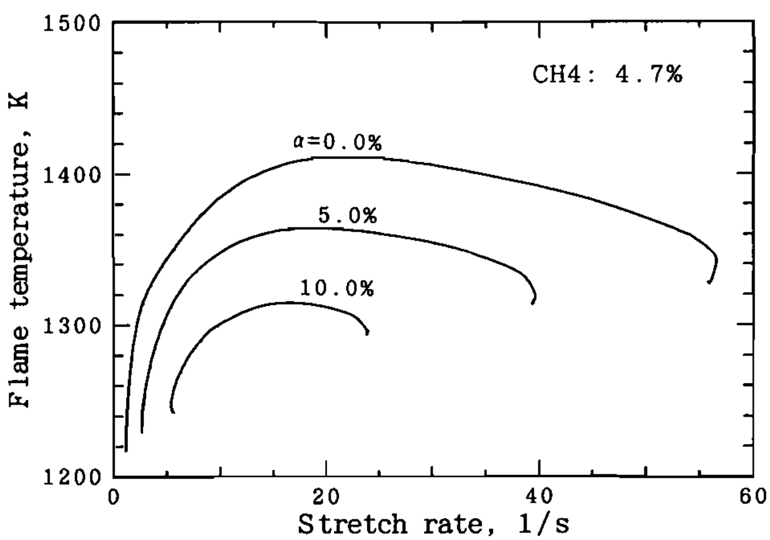

FIGURE 5 Flame temperatures, where α is the fraction of CO_2 added into air.

is longer near the radiation extinction limit than that near the stretch extinction limit. Thus the flame is more sensitive to the temperature at the stretch extinction limit than at the radiation extinction limit. This is why the reduction of stretch rate at the stretch extinction limit is greater than the increase of stretch rate at the radiation extinction limit, when the fraction of CO_2 added to air is increased. In fact, the radiation fraction defined in our previous study (Guo *et al.*, 1997) at the radiation extinction limit for the CO_2 fraction is 10.0% is less than that for corresponding CH_4/air flame due to the flame temperature reduction.

The C-shaped curves of CH_4/CO_2/air counterflow premixed flames for the fraction of CO_2 added to air being 0.0, 5.0 and 10.0%, respectively, are shown in Figure 6, where some experimental data are also shown. The velocity gradient, which is defined as U/L (where U is the velocity at the nozzle exit and L is the distance between the nozzle exit and the stagnation plane), in experiments corresponds to the value $2a$ (a is the stretch rate in the calculation). The experimental data by the present study were obtained under normal gravity condition, and the burner system and the mixture

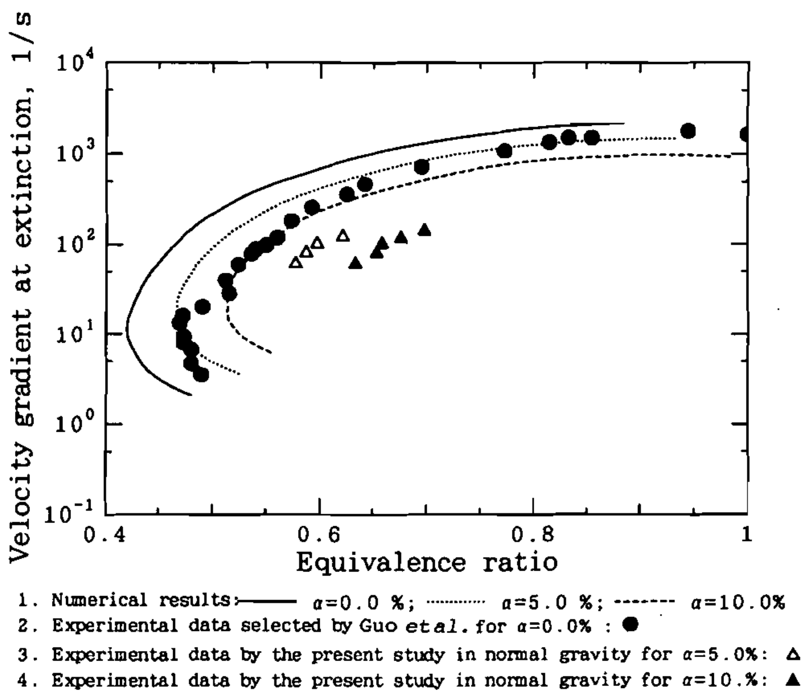

FIGURE 6 The C-shaped curves.

supply system are the same as those used in studies by Maruta *et al.* (1996) and Niioka *et al.* (1983), respectively. The C-shaped curves show that the flammable region of the CH_4/CO_2/air counterflow premixed flame becomes smaller with the increase of the fraction of CO_2 added to air. It can be found that the present numerical results appear the same qualitative tendency with the experimental results, although the present experiments were not extended to sufficiently low stretch rates due to the natural convection under normal gravity condition.

$CH_4/CO_2/O_2$ Flames

In order to eliminate the effect of heat capacity increase when CO_2 is added to air, numerical calculations were conducted for $CH_4/CO_2/O_2$ counterflow premixed flames. The adiabatic equilibrium flame temperature at every equivalence ratio is kept the same as that of CH_4/air flame by adjusting the ratio of CO_2 to O_2. The radiation model adopted is also the same as that used above.

Figure 7 shows the temperature variations *versus* stretch rate for four equivalence ratio $CH_4/CO_2/O_2$ flames. We can find that there are two extinction limits: the stretch extinction limit occurring at a higher stretch rate and the radiation extinction limit occurring at a lower stretch rate, for

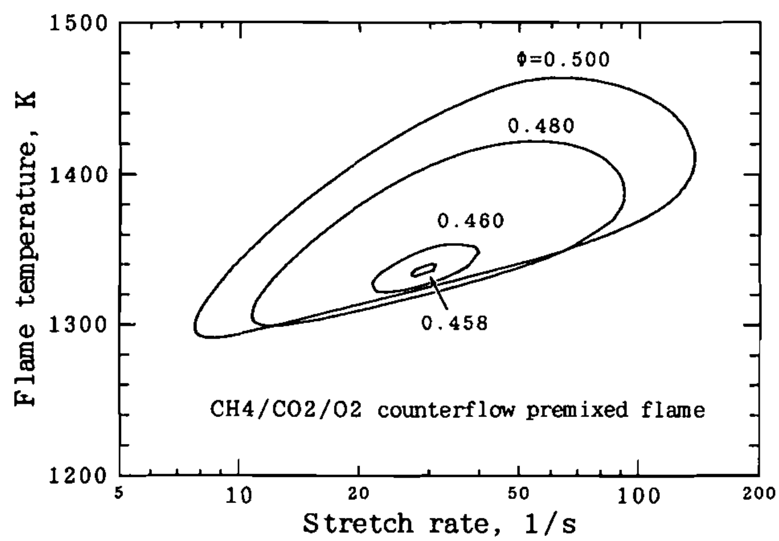

FIGURE 7 Flame temperature variations *versus* stretch rate.

every equivalence ratio flame, similar to those of CH_4/air and CH_4/CO_2/air flames.

Figure 8 compares the temperature variations between $CH_4/CO_2/O_2$ and CH_4/air flames for two equivalence ratios. It can be found that the stretch rates at radiation extinction limits of $CH_4/CO_2/O_2$ flames are much higher than those of CH_4/air flames for both equivalence ratios. This is due to the increase of radiation heat loss from $CH_4/CO_2/O_2$ flames, compared to CH_4/air flames. It is known that CO_2 is a species with stronger emitting ability, and thus radiation heat loss from the $CH_4/CO_2/O_2$ flame is more than that emitted from the CH_4/air flame if the flame temperature level is kept almost same. Therefore, due to radiation heat loss, $CH_4/CO_2/O_2$ flames extinguish at much higher stretch rates than CH_4/air flames when the stretch rate decreases.

However, it is interesting to note that the stretch rates at stretch extinction limits exhibit a different feature for two equivalence ratio flames. At the stretch extinction limit, the stretch rate of leaner $CH_4/CO_2/O_2$ flame ($\phi = 0.458$) is lower than that of CH_4/air flame, while the stretch rate of richer $CH_4/CO_2/O_2$ flame ($\phi = 0.480$) is higher than that of CH_4/air flame. It is believed that this is due to the effect of Lewis number, defined as the ratio of mixture thermal diffusivity to fuel mass diffusivity. The Lewis number of $CH_4/CO_2/O_2$ mixture is about 0.8, while the Lewis number of

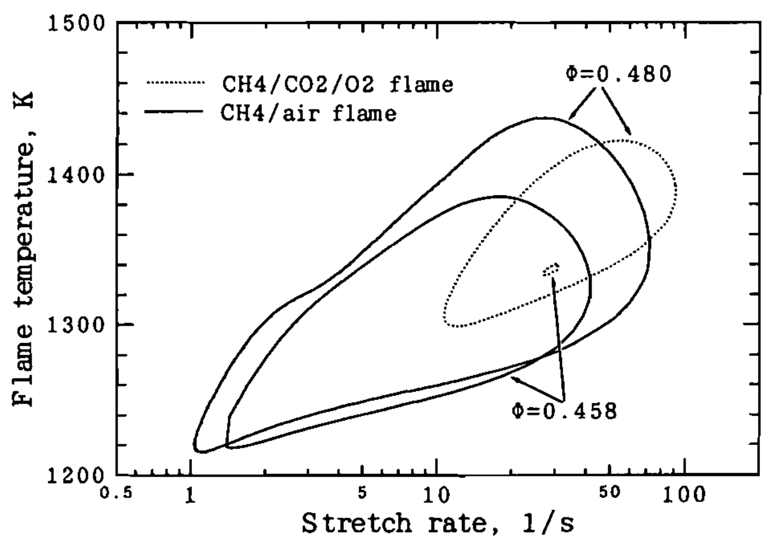

FIGURE 8 Flame temperature variations *versus* stretch rate.

CH_4/air mixture is about 0.98. When the equivalence ratio is 0.458, the flame can be sustained within the relatively lower stretch rate range, and thus the Lewis number effect is relatively weaker. So the $CH_4/CO_2/O_2$ flame extinguishes more easily than the CH_4/air flame due to radiation heat loss, and thus the stretch extinction limit of $CH_4/CO_2/O_2$ flame is a little lower than that of CH_4/air flame of the equivalence ratio being 0.458. On the other hand, the flame can be sustained within the relatively higher stretch rate range, and thus the Lewis number effect is relatively stronger when the equivalence ratio is 0.480. Therefore, the local equivalence ratio of reaction zone of the $CH_4/CO_2/O_2$ flame becomes greater at higher stretch rates due to the preferential diffusion of fuel than that of the CH_4/air flame, and so the $CH_4/CO_2/O_2$ flame can be sustained at a higher stretch rate than the CH_4/air flame, although they have the same adiabatic equilibrium flame temperature.

The C-shaped curves of both $CH_4/CO_2/O_2$ and CH_4/air flames are shown in Figure 9. As we discussed earlier, the stretch rates at the radiation extinction limit branch of $CH_4/CO_2/O_2$ flame are much higher than those of CH_4/air flame. The stretch rates at the stretch extinction limit branch of $CH_4/CO_2/O_2$ flame at very small equivalence ratio region are lower than those of CH_4/air flame. Therefore the concentration limit of $CH_4/CO_2/O_2$ flame is higher than that of CH_4/air flame. With the increase of equivalence ratio, the stretch

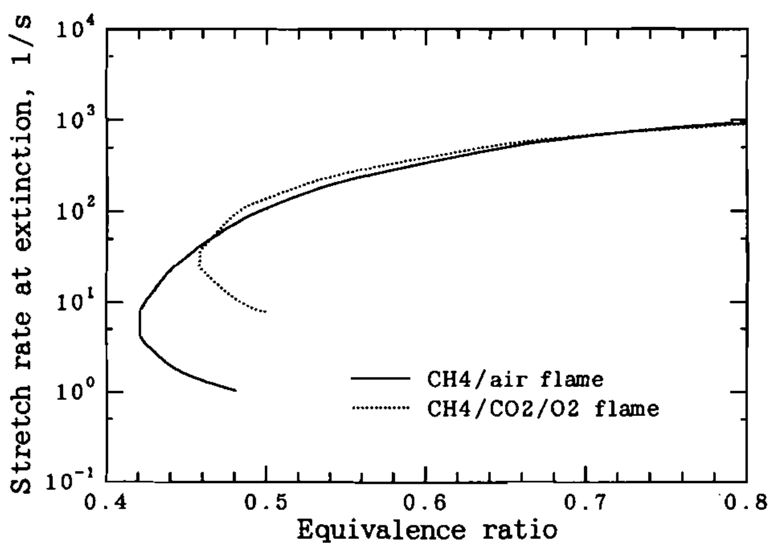

FIGURE 9 The C-shaped curves.

rates at stretch extinction limit branch of $CH_4/CO_2/O_2$ flame become higher than those of CH_4/air flame due to the effect of Lewis number. However, the difference between the stretch rates of two flame at the stretch extinction limit branches diminishes, even the stretch rates at stretch extinction limit branch of $CH_4/CO_2/O_2$ flame being a little lower than those of CH_4/air flame when the equivalence ratio further increases. This may be attributed to the fact that the local equivalence ratio of reaction zone begins to exceed unity due to the preferential diffusion of fuel for higher equivalence ratio flames. Consequently the Lewis number effect can no longer strengthen the combustion, and the stretch rates at stretch extinction limit branches become almost same for two flames, even the stretch rates at stretch extinction limit branch of $CH_4/CO_2/O_2$ flame being a little lower than those of CH_4/air flame at higher equivalence ratio region.

Of course, other thermal properties, such as heat conductivity, may also affect the extinction characteristics and cause the difference between $CH_4/CO_2/O_2$ and CH_4/air flames. Since here only the effect of radiation heat loss is concerned, discussions on the effects of other thermal properties are not the interest of this paper.

CONCLUSIONS

Numerical investigations of CH_4/CO_2/air and $CH_4/CO_2/O_2$ counterflow premixed flames by the detailed chemistry and transport properties were conducted in the present paper. A radiation model that can consider the radiation reabsorption was used in the calculation.

Results indicate that the radiation extinction limit still exists when radiation reabsorption is taken into account. Radiation reabsorption has little effect on the flame temperature and extinction limits of CH_4/air counterflow premixed flames, and thus almost has no effect on the C-shaped curve of CH_4/air counterflow premixed flames. However the reabsorption has an obvious influence on the flame temperature and extinction limits of CH_4/CO_2/air counterflow premixed flames, especially on the radiation extinction limit. Thus the reabsorption must be considered in the calculation of CH_4/CO_2/air counterflow premixed flames.

With the increase of CO_2 fraction in the CH_4/CO_2/air mixture, the flame temperature drastically decreases due to the increase of heat capacity of the mixture. Therefore the flammable region of CH_4/CO_2/air counterflow premixed flames becomes smaller when the fraction of CO_2 mixed into the air is increased.

For $CH_4/CO_2/O_2$ flames, the stretch rates at radiation extinction limit branch of C-shaped curve is much higher than those of CH_4/air flame due to radiation heat loss, and the concentration limit is also higher than that of CH_4/air flame.

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