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EFFECTS OF RADIATIVE EMISSION AND ABSORPTION ON THE PROPAGATION AND EXTINCTION OF PREMIXED GAS FLAMES

YIGUANG JU,¹ GORO MASUYA¹ AND PAUL D. RONNEY²

¹Department of Aeronautics and Space Engineering
Tohoku University
Aoba-ku, Sendai 980, Japan

²Department of Aerospace and Mechanical Engineering
University of Southern California
Los Angeles, CA 90089-1453, USA

Premixed gas flames in mixtures of CH₄, O₂, N₂, and CO₂ were studied numerically using detailed chemical and radiative emission-absorption models to establish the conditions for which radiatively induced extinction limits may exist independent of the system dimensions. It was found that reabsorption of emitted radiation led to substantially higher burning velocities and wider extinction limits than calculations using optically thin radiation models, particularly when CO₂, a strong absorber, is present in the unburned gas. Two heat loss mechanisms that lead to flammability limits even with reabsorption were identified. One is that for dry hydrocarbon-air mixtures, because of the differences in the absorption spectra of H₂O and CO₂, most of the radiation from product H₂O that is emitted in the upstream direction cannot be absorbed by the reactants. The second is that the emission spectrum of CO₂ is broader at flame temperatures than ambient temperature; thus, some radiation emitted near the flame front cannot be absorbed by the reactants even when they are seeded with CO₂. Via both mechanisms, some net upstream heat loss due to radiation will always occur, leading to extinction of sufficiently weak mixtures. Downstream loss has practically no influence. Comparison with experiment demonstrates the importance of reabsorption in CO₂-diluted mixtures. It is concluded that fundamental flammability limits can exist due to radiative heat loss, but these limits are strongly dependent on the emission-absorption spectra of the reactant and product gases and their temperature dependence and cannot be predicted using gray-gas or optically thin model parameters. Applications to practical flames at high pressure, in large combustion chambers, and with exhaust-gas or flue-gas recirculation are discussed.

Introduction

Flammability limit studies are important for assessing of fire safety in many environments and for determining the operation limits of combustion devices. Despite many years of study, the mechanisms of flammability limits of premixed gas flames are still not well understood. In particular, it has not been established whether "fundamental" limits exist independent of apparatus size and geometry. Many mechanisms cause apparatus-dependent extinction limits including flame stretch [1,2], buoyant convection [3,4] and heat losses to cold walls [5-7]. When these losses are eliminated by employing large combustion vessels at microgravity, heat loss via gas-radiation is probably the dominant extinction mechanism [8-11].

Nevertheless, it is unclear whether radiative loss causes fundamental limits because emitted radiation can be reabsorbed, a factor not considered by the theories [6,7] and computations [10,11]. Whether reabsorption is important is usually assessed by comparing the system dimension to the burned-gas

Planck mean absorption length (L_P), defined as the mean monochromatic absorption coefficient (κ) weighted by the Planck function:

$$L_P^{-1} = \int_0^\infty \kappa(u) G(u) du;$$
$$G(u) = \frac{15}{\pi^4} \frac{u^3}{e^u - 1}; u = hc\omega/kT \quad (1)$$

where h , c , ω , k , and T denote Planck's constant, light speed, wavenumber, Boltzmann's constant, and temperature, respectively. In the experiments in Refs. [8,9], the optical thickness $\tau = X/L_P$, where X is an apparatus dimension, was generally small (with exceptions noted later), consequently optically thin conditions (no reabsorption) applied. Theoretical studies of premixed-gas flames seeded with inert particles [12] predict that with reabsorption, net heat losses decrease, burning velocities (S_L) increase, and the flammability limit equivalence ratio (Φ_0) decreases compared with values without reabsorption.

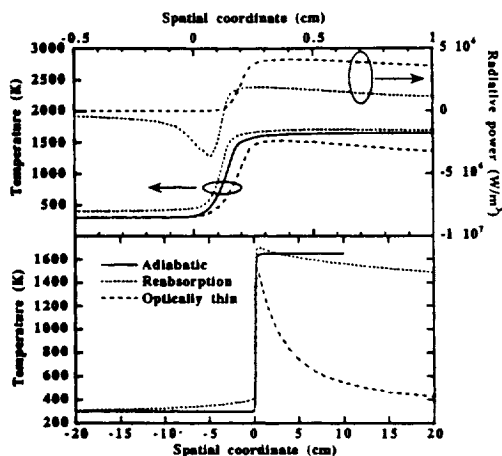


FIG. 1. Example of model predictions for $\Phi = 0.70$, $\gamma = 0.30$ (near the lean flammability limit for optically thin flames) for adiabatic, optically thin, and reabsorbing flames. The corresponding burning velocities are 7.9, 5.9, and 12.7 cm/s, respectively. The optical thickness $\tau = L_f/L_p$ is 2.39. Upper plot: temperature (T) and radiative power (Q_R), $-0.5 < x < 1.0$ cm; lower plot: temperature only, $-20 < x < 20$ cm.

Corresponding microgravity experiments in particle-laden methane-air mixtures [13] support these predictions. Gaseous flame behavior should be different from that of particle-laden flames because gases emit and absorb in spectral bands, whereas particles exhibit approximately gray-gas behavior. No computational studies of gaseous flames with detailed radiative emission/absorption models have been performed to test their effects on S_L and Φ_0 . Consequently, our goal is to model premixed-gas flame propagation with detailed radiative emission/absorption effects and compare results with experiments and theoretical predictions.

In addition to microgravity studies, modeling of premixed-gas flames with reabsorption is relevant to combustion at high pressures and in large furnaces because frequently $\tau > 1$. For example, at 40 atm, a typical pressure for premixed-charge internal combustion engines, $\tau \approx 1$ for cylinders of radius 4 cm. Similarly, $\tau > 1$ in atmospheric-pressure furnaces larger than 1.6 m. Moreover, significant amounts of absorbing CO_2 and H_2O are present in the unburned mixtures of combustion devices employing exhaust-gas or flue-gas recirculation.

Numerical Model

The energy and chemical species conservation equations for steady planar premixed-gas flames were solved using a CHEMKIN-based code [14] with arc-length continuation [15,16]. This code was

employed previously to model optically thin radiating counterflow flames [16]. For this study, radiative transport including both emission and absorption was computed using the statistical narrowband model with exponential-tailed inverse line strength distribution [17]. The radiative transfer equations were solved for wavenumbers between 150 and 9300 cm^{-1} with 25 cm^{-1} resolution using the S6 discrete ordinate method. Radiation parameters for CO_2 , H_2O , and CO were taken from Ref. [18]. CH_4 radiation was not included because the necessary spectral data were not available, but CH_4 radiation is minimal even for optically thin conditions [11]. Further details of the model, including accuracy considerations, are presented elsewhere [19]. Methane oxidation was modeled using an 18-species, 58-step chemical mechanism [14]. The spatial position (x) = 0 was defined as the location where $T = 325$ K for radiation-free flames. Except where noted, the upstream and downstream boundary locations were $x = -L_1 = -30$ cm and $x = +L_2 = +400$ cm, respectively. Upstream boundary conditions were ambient temperature (300 K), and composition with inflow velocity S_L and downstream boundary conditions were zero-gradient. Ambient-temperature blackbody walls were assumed at both boundaries.

Mixtures of $\text{CH}_4 + \{0.21\text{O}_2 + (0.79 - \gamma)\text{N}_2 + \gamma\text{CO}_2\}$ were examined, with γ varied to assess reabsorption effects by substituting emitting/absorbing CO_2 for radiatively inactive N_2 . Ambient H_2O was not considered because experiments at standard conditions allow at most 3% H_2O without condensation.

Results

Figure 1 shows typical computed results. For optically thin conditions, the volumetric radiative loss (Q_R) = $4\sigma(T^4 - T_0^4)/L_p$, where σ is the Stefan-Boltzmann constant and T_0 the ambient temperature, is always positive (upper plot). With reabsorption, Q_R is negative at $x < 0$ because some radiation emitted at high T ($x > 0$) is reabsorbed at lower T ($x < 0$). This decreases the net loss and preheats the unburned mixture so that, consistent with the theory in Ref. [12], the peak temperature (T_*) exceeds adiabatic (T_{ad}). With optically thin radiative loss, T decays downstream to T_0 (lower plot). With reabsorption, T still decays downstream (but on a much longer scale); thus, some net loss still occurs. All flames exhibit conventional convective-diffusive zones of thickness $\delta \approx \alpha/S_L \approx 0.1$ cm, where α is the temperature-averaged thermal diffusivity. The reabsorbing flame additionally exhibits a much longer upstream convective-radiative zone (lower plot) whose length is comparable to L_p (≈ 19 cm).

Fig. 2a shows that reabsorption effects in CH_4 -air mixtures ($\gamma = 0$) are minor because Φ_0 is reduced

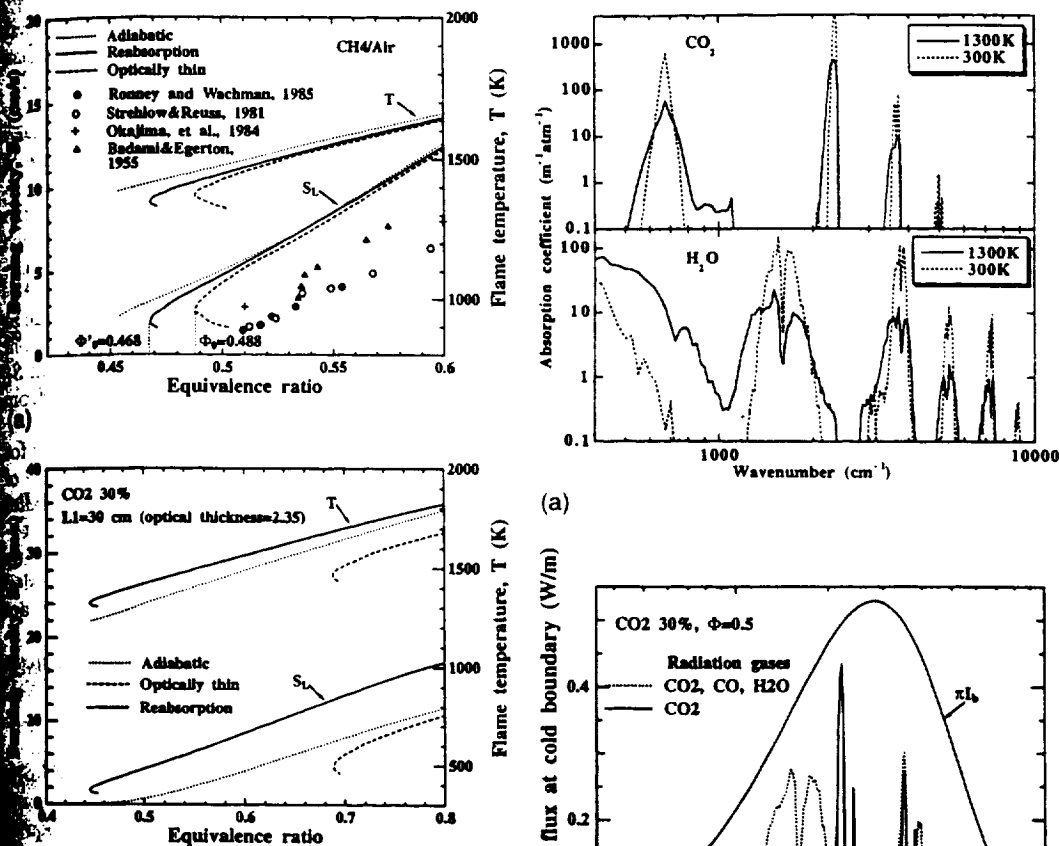


FIG. 2. Predicted burning velocities and peak flame temperatures under adiabatic conditions, with optically thin radiative losses and with a radiative model including reabsorption effects. In this and subsequent figures, the turning-point limit is shown, but for clarity, the unstable lower branch is omitted. (a) Lean CH_4 -air mixtures. (b) CH_4 -(0.21 O_2 + 0.49 N_2 + 0.30 CO_2) mixtures ($\gamma = 0.30$).

only 3.8% and, even with reabsorption $S_{L,r}$, is always less than the adiabatic burning velocity ($S_{L,ad}$). However, $S_{L,lim}$ is reduced by 25%, and $S_{L,ad}$ for the limit temperature decreases similarly (26%). For both optically thin and reabsorption models, $S_{L,lim}/S_{L,ad}$ is close to the theoretical prediction $e^{-1/2}$ [6,7]. A simple picture emerges from these results. Radiation from hot H_2O and CO_2 can escape only if it is emitted upstream, because downstream radiation will be reabsorbed by other H_2O and CO_2 molecules. Thus, reabsorption approximately halves the net heat loss. The theory for optically thin flames [6,7] predicts $S_{L,lim} = Q_R^{1/2}$; thus, if Q_R were halved, $S_{L,lim}$ should decrease 29%. Consequently, the classical quenching mechanism approximately applies in this case.

Flame characteristics are quite different with $\gamma =$

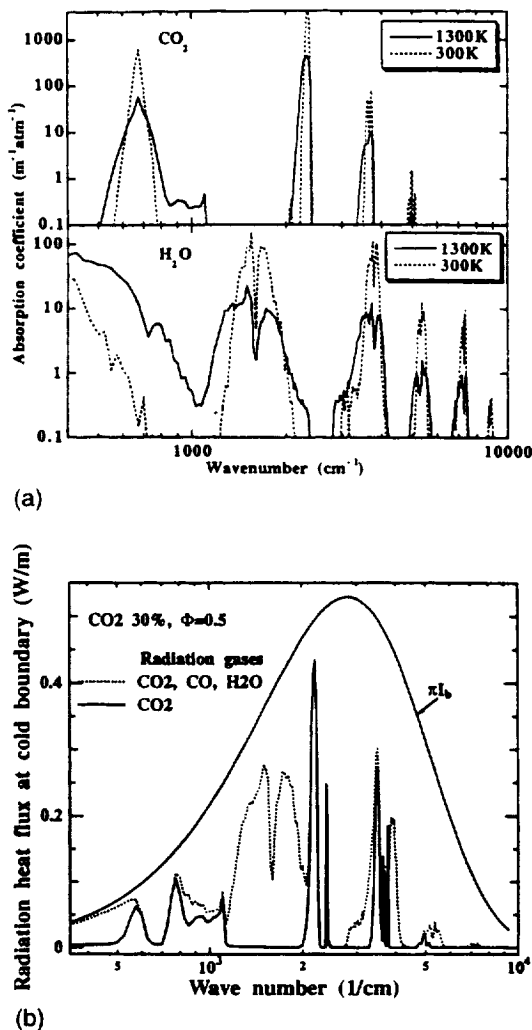


FIG. 3. Radiative properties of combustion gases as a function of wavenumber (ω). (a) Absorption coefficients $\kappa(\omega)$ for pure CO_2 and H_2O at 1300 K and 300 K. (b) Radiative heat flux at cold (upstream) boundary for $\phi = 0.50$, $\gamma = 0.30$ ($\tau = 2.39$) with reabsorption effects included. Note that the units on the vertical axis are W/m^2 per $\text{m}^{-1} = \text{W/m}$ (i.e., the radiant flux per unit wavenumber). Also shown is πI_b , where I_b is the Planck function for a blackbody at T^* for this flame (i.e., the maximum possible flux.)

0.30 (Fig. 2b). As with $\gamma = 0$, for optically thin radiation, S_L and T_* are lower than adiabatic values, but with CO_2 substitution, because of reabsorption, these quantities are significantly higher than adiabatic values. The effect on ϕ_{st} is substantial (0.682 for optically thin conditions vs. 0.442 with reabsorption). Nevertheless, flammability limits still exist.

Figure 3 elucidates two mechanisms that cause net

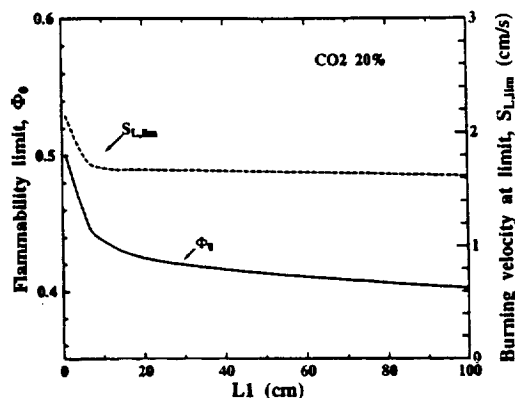


FIG. 4. Effect of upstream domain length (L_1) on flammability limit properties: equivalence ratio (Φ_0) and burning velocity ($S_{L,lim}$) for $\gamma = 0.20$. The corresponding flammability limit for optically thin conditions is $\Phi_0 = 0.62$.

heat loss and thus flammability limits even with reabsorption, designated I and II. Mechanism I is that $\kappa(\omega)$ is very different for H_2O and CO_2 (Fig. 3a); because H_2O is absent from the unburned gas, most H_2O radiation emitted upstream cannot be reabsorbed. Mechanism II is that $\kappa(\omega)$ for CO_2 is broader at T_* than T_0 (Fig. 3a); thus, some CO_2 radiation emitted upstream from temperatures near T_* cannot be reabsorbed farther upstream where $T \approx T_0$. Mechanism II can also occur for H_2O but not for the dry reactants studied here. Both mechanisms apply for arbitrarily large domains. The manifestation of these mechanisms is seen in the spectrally resolved radiative flux at the upstream boundary ($x = -L_1$) (Fig. 3b). The spectrum for $CO_2 + H_2O + CO$ is similar to the $CO_2 + H_2O$ spectra at 1300 K (Fig. 3a) less the CO_2 spectrum at 300 K, indicating that losses arise mainly from the differences between the burned-gas emission and unburned-gas absorption. H_2O emission comprises most of the loss (mechanism I), but some occurs from CO_2 emission (mechanism II).

These observations show that fundamental flammability limits due to radiative losses will exist in gaseous flames no matter how large the domain or what absorbing gases are present, because some radiation emitted from the high-temperature region cannot be reabsorbed by the unburned gases (mechanism II). These limits depend on the temperature effects on the absorption spectrum and cannot be predicted via simple mean absorption coefficients, as optically thin limits can. Of course, if sufficient quantities of inert particles, soot, or other quasi-graybody absorbers are present, complete reabsorption could occur.

Loss at the downstream boundary ($x = L_2$) is much less important for several reasons. A disappearing reactant (e.g., CH_4) can produce some loss

at $x = L_2$ via mechanism I, but CH_4 radiation was not considered, and the loss would be much less than the upstream H_2O and CO_2 loss because CH_4 disappears near $x = 0$ just as T rises to values where significant radiation could be emitted. Some downstream loss via mechanism II can occur because of the downstream temperature gradient, but the gradient and total decrease in T are much less than the corresponding upstream values, leading to much lower downstream loss. A third mechanism of radiative loss occurs at the downstream boundary because of the blackbody wall with $T = T_0$, but due to reabsorption, its influence is confined to the adjacent region of thickness L_p (≈ 18 cm). Thus, T_* and S_L are unaffected, as was verified by changing L_2 from 400 to 100 cm.

Losses via mechanisms I or II can occur only for wavenumbers where $\kappa(\omega)$ is nonzero on one side of the flame but changes to zero over a length smaller than the scale $[\kappa(\omega)]^{-1}$ over which reabsorption occurs. Changes occur on the scale δ for temperature and D_i/S_L for species i , where D_i is the diffusion coefficient. Because the Lewis numbers α/D_i are close to unity, δ is an appropriate scale for both species and temperature changes. Hence, these criteria become

$$\begin{aligned} \text{I: } \left| \frac{d\chi_i}{dx} \right| &> \left| \frac{d\chi_i}{d(1/\kappa(\omega))} \right| \Rightarrow \frac{d \ln(\kappa(\omega))}{d \ln(\chi_i)} \\ &> \kappa(\omega)\delta \Rightarrow \kappa(\omega)\delta < 1 \\ \text{II: } \left| \frac{dT}{dx} \right| &> \left| \frac{dT}{d(1/\kappa(\omega))} \right| \Rightarrow \kappa(\omega)\delta \\ &< \frac{d \ln(\kappa(\omega))}{d \ln(T)} \end{aligned} \quad (2)$$

where χ_i is the mole fraction of species i and the fact $\kappa \sim \chi_i$ has been used. Fig. 3b shows evidence of these criteria. The loss due to H_2O mimics $\kappa(\omega)$ except where $\kappa(\omega)$ is very large and reabsorption can occur within the convective-diffusive zone where χ_{H_2O} changes rapidly. For CO_2 , practically no loss occurs where $\kappa(\omega)$ is large at both 300 K and 1300 K (Fig. 3a), but substantial loss occurs for ω on the "wings" of these peaks where $d\kappa/dT$ is large.

The convective-radiative zone at $x < 0$ has a characteristic thickness $L_p \gg \delta$; thus, L_1 can influence reabsorbing flames drastically. Fig. 4 shows that Φ_0 decreases as L_1 increases because more reabsorption (thus lower net Q_R) occurs with larger L_1 . Because $S_{L,lim} \sim Q_R^{1/2}$, $S_{L,lim}$ also decreases. Significant reabsorption effects occur even for $L_1 = 1$ cm ($\tau = 0.054$) because CO_2 has absorption bands with $\kappa(\omega)$ up to $4000 \text{ m}^{-1} \text{ atm}^{-1}$ ($40 \text{ cm}^{-1} \text{ atm}^{-1}$) (Fig. 3a). For the limit conditions at $L_1 = 1$ cm, $\chi_{H_2O} = 0.19$; thus, significant absorption occurs on the scale $(0.190 \times 40 \text{ cm}^{-1})^{-1} = 0.13$ cm. This estimated

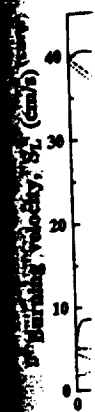


FIG. 5. Effect of upstream domain length (L_1) on burning velocity at limit ($S_{L,lim}$) for $\gamma = 0.20$.

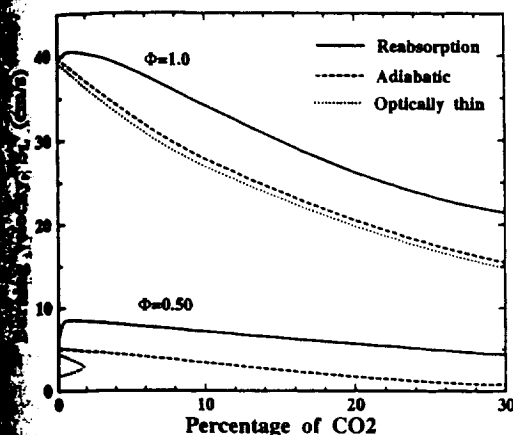


Fig. 5. Effect of substitution of CO_2 for N_2 on burning velocities under adiabatic conditions, with optically thin radiative losses, and with a radiative model including reabsorption effects.

minimum reabsorption scale is also seen in Fig. 1, which shows the negative (absorption) portion of Q_R decaying on a similar scale. Still, $L_1 > 100 \text{ cm}$ ($\tau > 4$) is required to obtain domain-independent results because the band "wings" have much smaller $\kappa(\omega)$ and thus longer absorption lengths. Consequently, τ provides only limited insight on flame properties with reabsorption.

Accurate determination of Q_R for small κ is difficult because for each 25 cm^{-1} spectral window, $\kappa(\omega)$ characterizes the sum of many individual absorption lines of widely varying strengths. For some individual lines but not others, absorption coefficients are factors of 10^{30} higher at 1300 K than at 300 K . Some lines overlap in ω -space while others are separated by transparent gaps that are required for net loss via mechanisms I or II to occur. Clearly, a single $\kappa(\omega)$ for each spectral window cannot capture all relevant details. Thus, although the reabsorption effects and flame characteristics proposed here are believed to be quantitative predictions will vary slightly depending on spectral resolution, line-shape model, and so on, especially for large L_1 .

Figure 5 shows that, for all radiation models, substituting CO_2 for N_2 has only minor effects on S_L in $\Phi = 1.0$ mixtures, except when γ is high enough to reduce T_{ad} and thus, $S_{L,ad}$ appreciably. In contrast, only 1% CO_2 substitution nearly doubles S_L for reabsorbing $\Phi = 0.5$ mixtures. There are two reasons why reabsorption effects are stronger at $\Phi = 0.5$. First, $\Phi = 0.5$ mixtures are much closer to the radiative extinction limit (note, for optically thin $\Phi = 0.5$ mixtures, very small amounts of CO_2 cause extinguishment due to increased Q_R) and thus benefit from the reduction in Q_R via reabsorption.

Second, $\Phi = 0.5$ mixtures have much higher Boltzmann numbers (B), which is a scaled ratio of blackbody emissive power at T_{ad} to total heat release rate and thus measures the potential for radiative preheating to increase S_L . For nonscattering media [12]

$$B = \frac{\beta \sigma (T_{ad}^4 - T_0^4)}{2 \rho_0 S_{L,ad} C_p T_{ad}}; \quad \beta = \frac{E}{RT_{ad}} \quad (3)$$

where β , E , R , ρ_0 , and C_p represent nondimensional activation energy, overall activation energy, gas constant, ambient density and specific heat, respectively. Values of B for $\gamma = 0$ mixtures are about 11.3 and 127 for $\Phi = 1.0$ and 0.50 , respectively; thus, reabsorption can increase S_L much more in $\Phi = 0.5$ mixtures.

Figure 6a shows that, for optically thin conditions, CO_2 substitution increases Φ_0 and $S_{L,lim}$, because the additional radiating CO_2 increases Q_R . With reabsorption, small amounts of CO_2 substitution actually decrease Φ_0 and $S_{L,lim}$ due to greatly reduced Q_R , whereas larger amounts increase Φ_0 slightly due to reduced T_{ad} . Fig. 6b shows that for optically thin conditions, $S_{L,lim}/S_{L,ad}$ is always close to the theoretical prediction $[6,7] e^{-1/2}$, whereas with reabsorption $S_{L,lim}/S_{L,ad}$ can be greater than 20.

Figure 7 shows comparisons of our computed effects of reabsorption on S_L to the theoretical prediction of Ref. [12] $S_L/S_{L,ad} \ln(S_L/S_{L,ad}) = B$ for gray gases. The theory requires that $(T_* - T_{ad})/T_{ad}$ be a small, $O(1/\beta)$ quantity, which is justified for our conditions (Fig. 2b). Agreement is poor because the computation allows substantial net spectral heat loss (Fig. 3b), which is not considered by the theory. Agreement is improved with H_2O and CO radiation suppressed (mechanism I eliminated) and still better with temperature broadening of $\kappa(\omega)$ also suppressed (mechanism II eliminated) so that the flame is practically adiabatic. Still, $S_L/S_{L,ad}$ is much lower than theoretical predictions because the theory assumes graybody radiation, whereas gases radiate only in certain spectral bands and thus accelerate S_L less than graybody radiators would. When gray-gas conditions are applied ($\kappa(\omega) = \text{constant} = 26 \text{ m}^{-1} = L_F^{-1}$), the agreement is more satisfactory, though calculated results are now above theoretical predictions, probably because the theory assumes constant thermodynamic and transport properties and single-step chemistry. Thus, our model is believed to capture the essential elements of flame propagation with graybody reabsorption plus the nuances of spectrally radiating gases.

Comparison with Experiment

For CH_4 -air flames (Fig. 2a), comparison between computation and microgravity experiments in spherically expanding flames [20,21] and tubes [22] and earth-gravity experiments specially designed for low

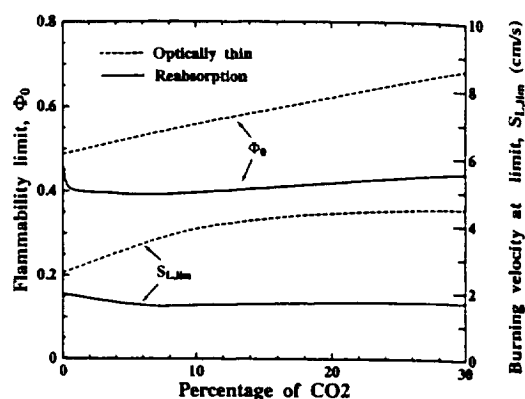
S_L [23] is more favorable without reabsorption because optically thin conditions are applied to these experiments. The discrepancy between computation and experiment is significant even though the chemistry and transport models employed predict S_L away from the limits quite well [24]. There is other evidence that different chemical models capable of predicting burning velocities of near-stoichiometric flames yield varying predictions of near-limit properties [25].

The only relevant S_L data that may exhibit optically thick conditions are microgravity spherically expanding flame experiments [9,26] in $(\text{CH}_4 + 4\text{O}_2) - \text{CO}_2$ mixtures and counterflow twin-flame experiments [27] in $\text{CH}_4 - (0.21\text{O}_2 + 0.79\text{CO}_2)$ mixtures ($\gamma = 0.79$). Neither configuration corresponds to our planar flames, but comparisons are nonetheless considered useful. To compare with Refs. [9,26], we chose $L_1 = L_2 = 6$ cm, which corresponds to a flame radius half the vessel radius. By this radius, S_L was steady, yet the pressure rise in the constant-volume vessel was small ($<10\%$). To compare with Ref. [27], we chose $L_1 = L_2 = 0.35$ cm, which corresponds to twin flames located midway between the nozzle exits and stagnation plane for 1.4-cm nozzle separation. For both experiments, ambient values of L_p are about 5.4 cm, but even for $L_1 = L_2 = 0.35$ cm, some reabsorption is anticipated (Fig. 3a).

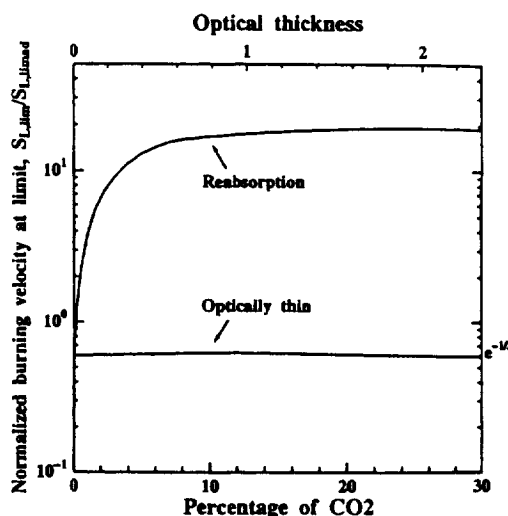
For the configuration of Refs. [9,26], the optically thin model clearly overpredicts the limit fuel concentration (by 13%) and $S_{L,\text{lim}}$ (by 350%); thus, reabsorption extends this limit. With reabsorption, $S_{L,\text{lim}}$ is predicted well, indicating that the net loss is correctly predicted, but the limit fuel concentration is underpredicted (by 16%), perhaps because in spherical geometry the radiative flux divergence causes less radiative preheating than would occur in planar geometry. For the configuration of Ref. [27], no numerical solutions could be obtained for adiabatic or optically thin conditions with $L_1 = L_2 = 0.35$ cm; this might explain why in Ref. [27] numerical predictions of S_L could not be obtained. In Ref. [27], results were extrapolated to zero strain to estimate the planar S_L . Although the accuracy of this method for reabsorbing conditions has not been established, with reabsorption the computed and experimental values of S_L agree moderately well (Fig. 8). Therefore, even for these small-scale flames, reabsorption effects may be important.

Conclusions

The effects of radiation on premixed-gas flames were studied using a detailed emission-absorption model for H_2O , CO_2 , and CO . It was found that while reabsorption can increase burning velocities and extend flammability limits considerably, fundamental limits independent of the system dimensions



(a)



(b)

FIG. 6. Effect of substitution of CO_2 for N_2 on flammability limit properties with optically thin radiative losses and with a radiative model including reabsorption effects. (a) Equivalence ratio (Φ_0), burning velocity ($S_{L,\text{lim}}$), and peak flame temperature (T^*). (b) Ratio of $S_{L,\text{lim}}$ to the burning velocity of an adiabatic flame in the same mixture ($S_{L,\text{ad}}$).

exist due to the nature of gas radiation, specifically, (1) differences between the spectral characteristics of reactants and products and (2) temperature broadening of the emission/absorption spectra. The results agree well qualitatively and in some cases quantitatively with theory and experiments.

In future work, we will examine stationary "flame balls," since modeling of recent space experiments [25] suggests dominant reabsorption effects in some cases. The spherically expanding flame configuration will be studied to compare with the microgravity experiments already cited [9,26]. The effects of elevated pressures will be examined because of their

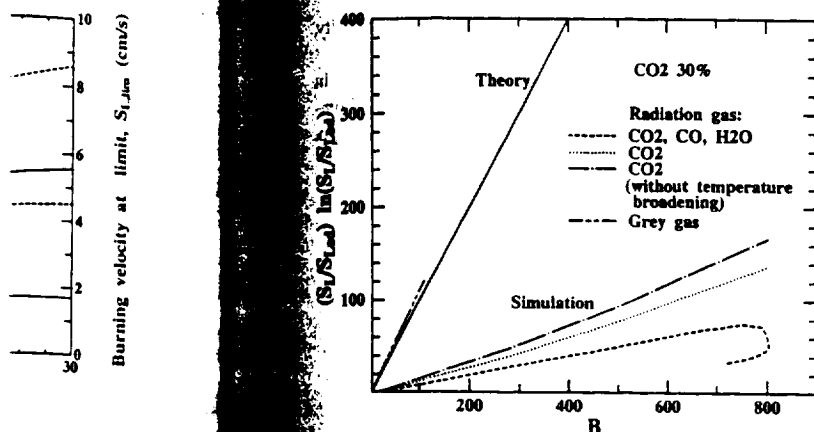


FIG. 7. Comparison of theoretical predictions [12] of reabsorption effects on S_L to numerical results with all radiation included, with CO_2 radiation only, with CO_2 radiation only with the temperature broadening of the absorption spectrum artificially suppressed, and with $\kappa(\omega) = \text{constant} = 26 \text{ m}^{-1}$ to simulate a gray gas.

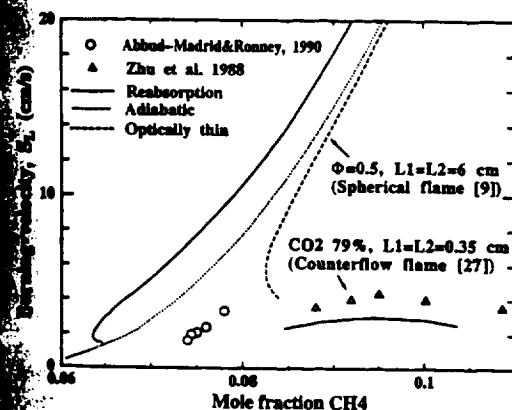


FIG. 8. Comparison of numerical predictions of S_L with experiments on microgravity spherically expanding flames ($\text{CH}_4 + 4\text{O}_2$)- CO_2 mixtures [9] and counterflow twin-flame experiments in CH_4 -air mixtures with $\gamma = 0.79$ (all H_2 replaced by CO_2) [27].

relevance to the internal combustion engine and gas turbine flames. Collisional broadening of the absorption spectrum will likely be important in these cases. Finally, the effects of exhaust-gas or flue-gas recirculation will be examined via computations at elevated temperatures with CO_2 and H_2O addition.

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