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Effects of radiation absorption on spherical flame propagation and radiation-induced uncertainty in laminar flame speed measurement

Zheng Chen*

SKLTCS, Department of Mechanics and Engineering Science, College of Engineering, Peking University, Beijing 100871, China

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Abstract

Outwardly propagating spherical flames are popularly used to measure the laminar flame speed, especially for high pressure conditions. Since radiation always exists in spherical flame experiments, the accuracy of laminar flame speed measurement is inherently affected by radiation. In this study, the radiation-induced uncertainty in laminar flame speed measurement was investigated numerically. We focused on CO_2 diluted mixtures in which the radiation absorption effects are important. The outwardly propagating spherical flames of different CO_2 diluted mixtures at a broad range of pressure up to 25 atm were simulated. Different fuels (hydrogen, methane, dimethyl ether and iso-octane) with different amounts of CO_2 dilution were considered and detailed chemistry was included in simulation. Two radiation models were used: one is the optically thin model considering only radiation emission and the other is the statistical narrow band model considering both radiation emission and absorption. The effects of radiation absorption on spherical flame propagation and radiation-induced uncertainty in laminar flame speed measurement were quantified through comparison among results predicted by these two radiation models. It was found that for CO_2 diluted mixtures, radiation absorption has great impact on spherical flame propagation: it greatly reduces the radiation-induced thermal and flow effects. The influence of radiation absorption was show to be stronger at higher pressure. When only radiation emission is considered and radiation absorption is neglected, the radiation-induced uncertainty in laminar flame speed measurement is substantially over-predicted for CO_2 diluted mixtures. When radiation absorption is included, the radiation-induced uncertainty in laminar flame speed measurement is nearly negligible (within 2.5%) for all the CO_2 diluted mixtures considered in this study.

Keywords: Laminar flame speed; Uncertainty; Radiation absorption; Spherical flame

1. Introduction

Laminar flame speed, S_u^0, is an important target for chemical mechanism validation [1]. Several experimental approaches have been developed to

* Tel.: +86 10 62766232.
E-mail addresses: cz@pku.edu.cn, chenzheng@coe.pku.edu.cn

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measure S_u^0. Among them, the outwardly propagating spherical flame (OPF) method (e.g., [2–4]) is popularly used, especially at high pressure. This method is investigated in the present work. Since the sensitivity of S_u^0 to chemical kinetics is relatively low, great attention has been recently paid to the accuracy of S_u^0 measurement ([1,5] and references therein). Quantification of uncertainty in S_u^0 measurement is extremely important when it is used to constrain the uncertainty of chemical models [1,5]. In this study, the radiation-induced uncertainty in S_u^0 measurement using the OPF method is investigated with the emphasis on the influence of radiation absorption.

Since radiation always exists in spherical flame experiments, S_u^0 measured from the OPF method is inherently affected by radiation. Theoretical studies (e.g., [6–8]) have demonstrated that radiation has great impact on spherical flame propagation under certain conditions. Unfortunately, it is difficult to accurately quantify the radiation-induced uncertainty in S_u^0 measurement since radiation depends nonlinearly on temperature and mixture properties.

As summarized in [9], there are several studies (e.g., [10–12]) on radiation-induced uncertainty in S_u^0 measured from the OPF method. Yu et al. [9] proposed an empirical correlation to quantify such radiation-induced uncertainty. However, the correlation does not work for mixtures diluted with CO_2, which has strong radiation absorption. In the literature, there are many studies (e.g., [13–17]) on S_u^0 measurement of CO_2 diluted mixtures due to their application in exhaust gas recirculation and oxy-fuel combustion systems. However, in most of these studies, the influence of radiation on the accuracy of S_u^0 measurement was not quantified. Santner et al. [11] studied the radiation-induced uncertainty of S_u^0 data measured for diluted H_2/O_2 mixtures [17]. However, they only used the optically thin model and did not consider radiation absorption. It will be shown here that the radiation-induced uncertainty was in fact over-predicted in [11]. More recently, Sohn et al. [18] have investigated the radiation effects on the uncertainty of S_u^0 determination for mixtures with $CO/CO_2/H_2O$ dilutions. They found that the optically thin model over-predicts the radiation effects and proposed two radiation corrections for S_u^0 determination from the OPF method. However, the corrections are difficult to be conducted since the required information on flow speed and density of burned gas is difficult to be obtained in experiments. Currently, for CO_2 diluted mixtures, it is still not clear how and to what extend radiation absorption affects the radiation-induced uncertainty in S_u^0 measured from the OPF method.

Furthermore, S_u^0 measurement at high pressure has recently received great attention due to its application in chemical mechanism development (e.g., [17]). However, few studies investigated the radiation-induced uncertainty in S_u^0 measurement at high pressure. At higher pressure, the flame propagation speed become smaller and the radiative heat flux become larger. Therefore, the radiation-induced uncertainty is expected to increase with pressure. However, for CO_2 diluted mixtures, radiation absorption also increases with pressure and thereby it can reduce the radiation-induced uncertainty. Therefore, it is still not clear how the radiation-induced uncertainty changes with pressure for CO_2 diluted mixtures.

Based on the above-mentioned considerations, the objective of this study is to quantify the effects of radiation absorption on spherical flame propagation and radiation-induced uncertainty in S_u^0 measurement at normal and high pressures. Simulations for different CO_2 diluted mixtures at a broad range of pressure from 1 to 25 atm were conducted and the influence of radiation absorption was quantified through comparison among results predicted by different radiation models. It is also valuable to assess the radiation-absorption effects on the flame propagation velocity in general case using simplified model. However, such theoretical analysis only provides general qualitative conclusions, and it cannot provide quantitative results since detail chemistry and spectral-dependent radiation cannot be considered in a simple model. Therefore, effects of radiation absorption on spherical flame propagation and radiation-induced uncertainty in S_u^0 measurement were quantified in this study through detailed simulation rather than theoretical analysis.

2. Numerical methods and specifications

One-dimensional simulation of outwardly propagating spherical flames was conducted and different radiation models were considered. The in-house code A-SURF [10,19,20] was used in simulation. A-SURF solves the conservation equations for a multi-species reactive flow using the finite volume method. A-SURF was used in previous studies on ignition and spherical flame propagation (e.g., [21–23]). The details on governing equations, numerical methods and code validation of A-SURF were presented in [10,19,20].

To quantify the influence of radiation absorption on spherical flame propagation and radiation-induced uncertainty in S_u^0 measurement, we compared results predicted by two radiation models: the optically thin model (denoted by 'OTM') considering only radiation emission and the statistical narrow band model (denoted by 'SNB') considering both radiation emission and absorption. For OTM, radiation emission from CO_2, H_2O, CO, and CH_4 was considered and the Planck mean coefficients in [24] were used. For SNB, the radiative transport was calculated using a fitted

Table 1
Summary of mixtures and pressure range.

No.	Mixture	Pressure	Ref.
#1	$0.083CH_4+0.275O_2+0.183He+0.459CO_2$ (i.e., $\phi=0.6$)	1–20 atm	[13]
#2	$0.051CH_3OCH_3+0.191O_2+0.279He+0.279N_2+0.2CO_2$ (i.e., $\phi=0.8$)	2–20 atm	[14]
#3	Fuel/air/CO_2; fuel = CH_4, CH_3OCH_3 or iC_8H_{18}; $\phi=1.0$; 0–20% vol. CO_2 in the whole mixture	1 atm	[15]
#4	$CH_4/O_2/CO_2$, $Z=40$–70%, $\phi=1.0$	1 atm	[16]
#5	$CH_4/O_2/CO_2$, $Z=0.6$, $\phi=0.5$–1.6	1 atm	[16]
#6	$H_2/O_2/CO_2$, $T_f=1500$, 1600, 1700, 1800 K, $\phi=2.5$	1–25 atm	[17]

Note: Z is defined as the volumetric ratio of CO_2 in the O_2/CO_2 mixture. The molar fractions of H_2, O_2 and CO_2 in mixtures #6 were specified in the Supplementary Document of [17].

statistical narrow-band correlated-k (FSNB-CK) method [13]. Besides, the adiabatic case (denoted by 'ADI') neglecting radiation was also simulated for comparison.

Our previous study [9] was focused on fuel/air mixtures without CO_2 dilution, for which the radiation absorption effects were shown to be almost negligible (see Fig. 3 in [9]). Here we studied mixtures diluted with different amounts of CO_2 which has strong radiation absorption. As summarized in Table 1, six mixtures were considered here. The laminar flame speeds of all these mixtures were measured by the propagating spherical flame method in the literature [13–17]. Therefore, it is necessary to understand how these S_u^0 measurements were affected by radiation absorption. Detailed chemistry was considered in simulation: GRI-Mech 3.0 [25] for methane, and the mechanisms of Zhao et al. [26], Chaos et al. [27] and Burke et al. [28] for dimethyl ether (DME), iso-octane and hydrogen, respectively. The CHEMKIN packages [29] were included in A-SURF to calculate the reaction rates and the mixture-averaged model was used to calculate mass diffusivity.

In order to diminish the effects of pressure rise and wall confinement [10,30] on spherical flame propagation, a large chamber radius of 50 cm was considered in simulation. Effects of instability and buoyancy were not included in 1D simulation. Therefore, only radiation effect appears and it can be readily quantified. We only considered spherical flames with radii below 2.5 cm since such flames were used in experiments (e.g., [13–17]) measuring S_u^0. For the linear extrapolation to be described in Section 3.2, spherical flames with radii in the range of $1.0 \le R_f \le 2.0$ cm were used so that the effects of ignition [21] and compression [10,30] on S_u^0 measurement can be minimized.

The premixture was initially static with the temperature of 298 K. A broad range of initial pressure was considered for mixtures #1, #2 and #6, as indicated in Table 1. Zero flow speed and zero gradients of temperature and mass fractions were enforced at both the center and wall boundaries. A small hot kernel (1–2 mm in radius) was used to initialize the spherical flame propagation from the center. Adaptive mesh refinement was used to accurately and efficiently resolve the propagating flame front. At high pressure of 25 atm, the smallest mesh size was reduced to 1 μm so that the reaction zone was covered by more than 15 grids. Grid independence was ensured for the simulation results presented in this study.

3. Results and discussion

The influence of radiation absorption was quantified through comparison between results predicted by OTM and SNB models since OTM only considers radiation emission while SNB includes both radiation emission and absorption.

3.1. Effects of radiation absorption on spherical flame propagation

Fig. 1 compares the temperature and flow speed distributions predicted by ADI, OTM and SNB for mixture #1 at $T_u = 298$ K and $P = 10$ atm. When there is no radiative loss, the temperature of burned gas is close to the adiabatic flame temperature T_f, and the burned gas is static (i.e., $u = 0$). When only radiation emission is considered as in OTM, the burned gas temperature is shown to continuously decrease as the flame propagates outwardly and the flame temperature is about 50 K lower than T_f. Moreover, it is observed in Fig. 1(b) that the flow speed of burned gas becomes negative and its magnitude continuously increases during the flame propagation. This inward flow is caused by radiation cooling of burned gas as explained in [9,10]. Therefore, radiative loss slows down spherical flame propagation in two ways: (1) it reduces flame temperature and thereby weakens the flame; and (2) it induces inward flow and thus inhibits flame propagation. These were respectively referred to as radiation-induced thermal and flow effects in [10].

When radiation absorption is considered as in the SNB model, Fig. 1 shows that both flame

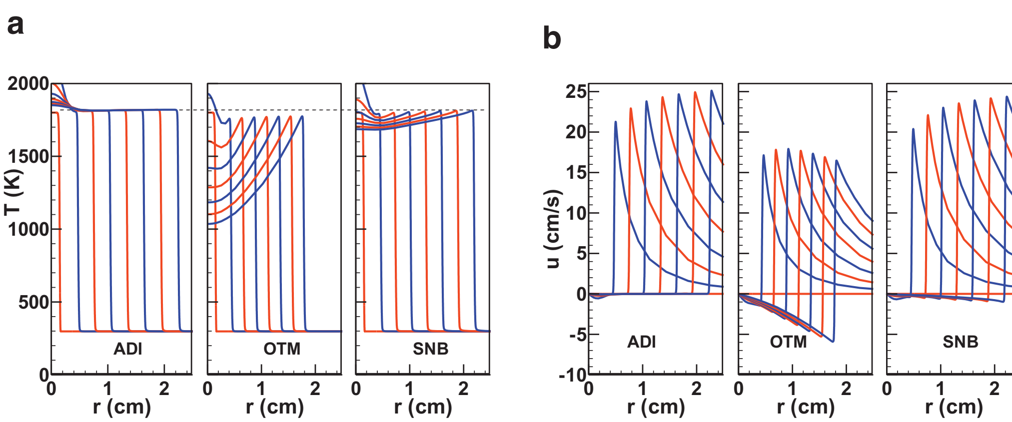

Fig. 1. Distributions of (a) temperature and (b) flow speed for a propagating spherical flame in mixture #1 at $T_u = 298$ K and $P = 10$ atm. The distributions are at consecutive equidistant instants of time: $t = 0$, 10, 20, 30, 40, 50, 60, 70 ms.

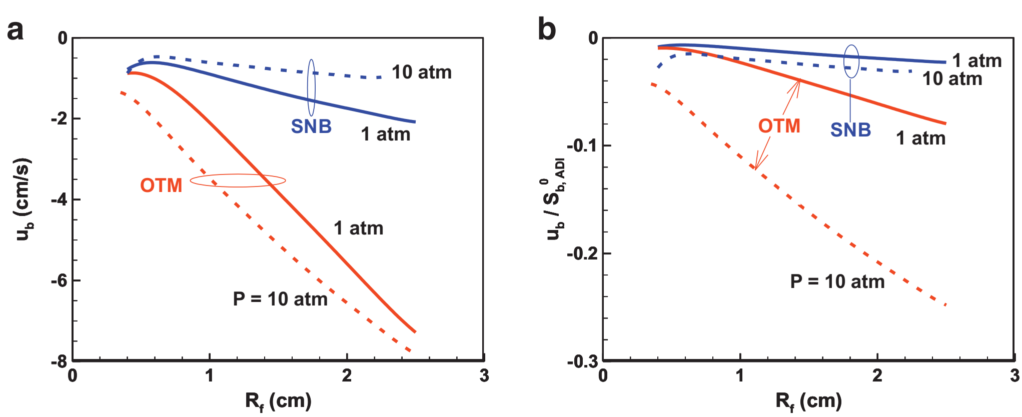

Fig. 2. Change of burned gas speed with flame radius for mixture #1 at 1 and 10 atm.

temperature and flow speed of burned gas are close to those of the adiabatic case. This demonstrates that radiation absorption can greatly reduce the radiation-induced thermal and flow effects. Therefore, radiation absorption has great impact on spherical flame propagation in CO_2 diluted mixture.

To quantify the radiation-induced flow effect, Fig. 2(a) plots the burned gas speed, u_b, as a function of flame radius for mixture #1 at 1 and 10 atm. u_b is the flow speed at the position where 99.9% of total heat release occurs. It is nearly the same as the minimum flow speed of burned gas (the relative difference is within 1%). Fig. 2(a) shows that $|u_b|$ increases with flame radius and thereby the radiation-induced flow effect becomes stronger at larger flame radius. Moreover, at P = 1 atm, u_b predicted by SNB is around one quarter of that by OTM, indicating that the radiation-induced flow effect is significantly reduced by radiation absorption. At higher pressure of 10 atm, the difference between u_b predicted by OTM and SNB becomes even larger since the radiation absorption heat flux is nearly proportional to pressure. Since the flame speed is smaller at higher pressure, the relative influence of radiation-induced flow on spherical flame propagation becomes much stronger at higher pressure, as demonstrated in Fig. 2(b). Therefore, the influence of radiation absorption on spherical flame propagation becomes stronger at higher pressure.

3.2. *Effects of radiation absorption on uncertainty in laminar flame speed measurement*

Fig. 3 shows the flame propagation speed S_b as a function of stretch rate K for mixture #1 at 1 and 10 atm. For propagating spherical flames, we have $S_b = dR_f/dt$ and $K = (2/R_f)(dR_f/dt)$, in which the flame radius, R_f, was determined as the location of peak heat release rate in simulation. The results do not change when other definitions/isotherms (e.g., the maximum density gradient) were used to deter-

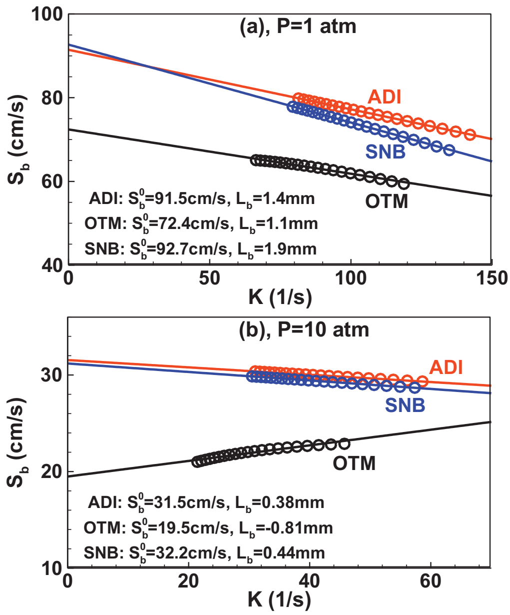

Fig. 3. Flame propagation speed as a function of stretch rate for mixture #1. The symbols are data for spherical flames with radii in the range of $1.0 \le R_f \le 2.0$ cm; and the solid lines stand for linear extrapolation.

mine R_f. Fig. 3 indicates that there is large difference between S_b predicted by ADI and OTM and that the relative difference increases greatly with pressure. Such difference is caused by the radiation-induced thermal and flow effects. It is seen that the difference between S_b predicted by ADI and SNB is much smaller than that between S_b predicted by ADI and OTM. This further demonstrates that the radiation-induced thermal and flow effects are substantially reduced by radiation absorption.

In $S_u{}^0$ measurement using propagating spherical flames, the burned gas is usually assumed to be static. Therefore, S_b is considered as the stretched flame speed relative to burned gas. Linear extrapolation between S_b and K based on the correlation of $S_b = S_b{}^0 - L_b K$ yields the unstretched laminar flame speed $S_b{}^0$ and Markstein length L_b, both relative to burned gas. The laminar flame speed $S_u{}^\circ$ can be obtained as: $S_u{}^0 = \sigma S_b{}^0$, where σ is the density ratio between burned and unburned mixtures. The values of $S_b{}^0$ and L_b from different radiation modes are presented in Fig. 3. When radiation absorption is neglected as in OTM, the relative difference between $S_b{}^0$ predicted by ADI and OTM is 21% and 38% for $P = 1$ and 10 atm, respectively. However, when radiation absorption is considered, the relative difference between $S_b{}^0$ predicted by ADI and SNB is within 2% for both $P = 1$ and 10 atm. Therefore, for mixture #1 containing 45.9% vol. CO_2, the radiation-induced uncertainty in $S_u{}^0$ measurement is substantially over-predicted by OTM. After considering radiation absorption, the radiation-induced uncertainty is in fact nearly negligible compared to uncertainties caused by other factors (for examples, mixture preparation, ignition, instability, confinement, extrapolation, etc.) summarized in [5].

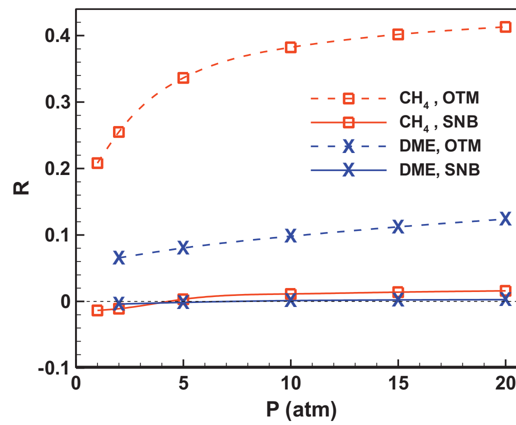

Fig. 4. Radiation-induced relative laminar flame speed reduction at different pressures for mixtures #1 (CH_4) and #2 (DME).

It is noted that Fig. 3(a) also indicates that $S_b{}^0$ predicted by SNB is slightly higher than that by ADI. This is because radiation absorption increases the temperature of unburned gas and thereby the laminar flame speed considering radiation absorption can be higher than that of the adiabatic case [31].

The radiation-induced relative reduction in laminar flame speed can be quantified by R defined as

$$R = 1 - S^0_{b,\text{radiative}} / S^0_{b,\text{ADI}} \qquad (1)$$

in which $S^0_{b,\text{radiative}}$ is predicted by OTM or SNB. R is equal to the radiation-induced uncertainty in laminar flame speed measured from propagating spherical flames. The influence of radiation absorption on R can be assessed by comparison between R predicted by OTM and SNB.

Fig. 4 shows the results for mixtures #1 ($CH_4/O_2/He/CO_2$ with 45.9% vol. CO_2) and #2 ($DME/O_2/He/N_2/CO_2$ with 20% vol. CO_2) at a broad range of pressure up to 20 atm. When radiation absorption is not considered as in OTM, R increases with pressure and it is above 20% and 6% respectively for mixtures #1 and #2. However, R predicted by SNB is shown to be within 2% for both mixtures at $P = 1$–20 atm. Therefore, the uncertainty in $S_u{}^0$ measurement is exaggerated by OTM (in which the radiation absorption is not considered) and it is greatly reduced by radiation absorption. Consequently, for mixtures #1 and #2 with large amount of CO_2 dilution, the radiation-induced uncertainty in $S_u{}^0$ measurement is negligible since it is much smaller than the un-

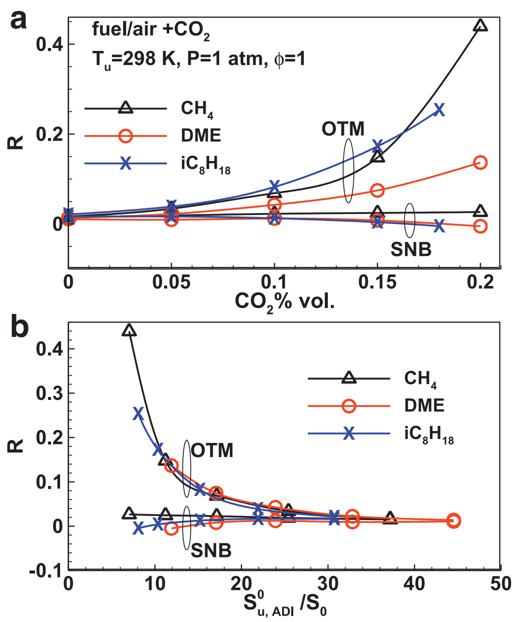

Fig. 5. Relative laminar flame speed reduction, R, as a function of (a) CO_2 dilution ratio and (b) normalized flame speed ($S_0 = 1$ cm/s) for mixtures #3.

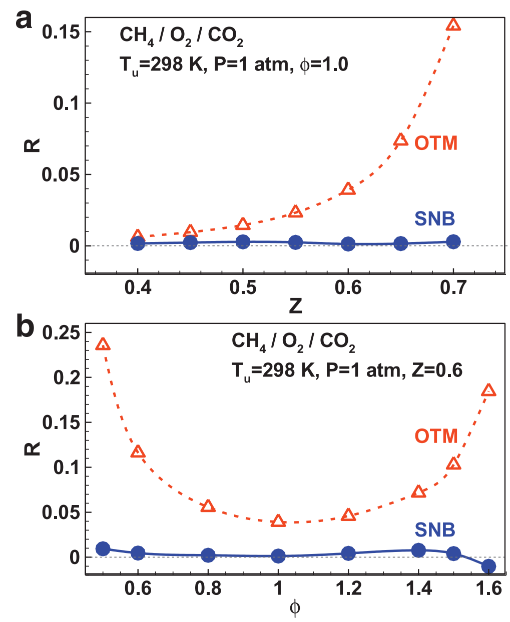

Fig. 6. Relative laminar flame speed reduction, R, as a function of (a) volumetric fraction of CO_2 in O_2/CO_2 mixture (i.e., mixture #4) and (b) equivalence ratio (i.e., mixture #5).

certainty inducted by other factors such as mixture composition and extrapolation [5].

Since many experiments (e.g., [15]) were conducted to measure S_u^0 of fuel/air mixtures with different amounts of CO_2 dilution, we investigated the influence of radiation absorption on R for such kind of mixtures (mixture #3 in Table 1). The results were plotted in Fig. 5 for CH_4, DME and iC_8H_{18}. Fig. 5(a) shows that when radiation absorption is not considered as in OTM, R increases greatly with CO_2 dilution and it is 15%, 7.5% and 17% respectively for CH_4, DME and $iC_8H_{18}$with 15% vol. CO_2 dilution. This indicates that the radiation-induced uncertainty predicted by OTM cannot be neglected. However, when radiation absorption is considered, the radiation-induced uncertainty predicted by the SNB model is within 2% and thereby it is negligible. Again, this demonstrates that the radiation-induced uncertainty in S_u^0 measurement can be substantially reduced by radiation absorption.

Fig. 5(a) indicates that R predicted by OTM is the smallest for DME. This is mainly due to the fact that the laminar flame speed of DME is higher than that of CH_4 and iC_8H_{18}at the same amount of CO_2 dilution. In Fig. 5(b) we plotted R as a function of normalized flame speed. It is seen that the results of different fuels fall almost on the same line, indicating that R correlates well with the laminar flame speed [9].

In oxy-fuel combustion, CO_2 dilution is usually used to reduce flame temperature. The laminar flame speeds of CO_2-diluted, methane oxy-fuel mixtures (mixtures #4 and #5 in Table 1) were measured by Xie et al. [16]. Therefore, here we examined the radiation absorption effects on the radiation-induced uncertainty in S_u^0 measurement for such oxy-fuel mixtures. Fig. 6(a) depicts the radiation-induced uncertainty, R, as a function of Z (defined as the CO_2 volumetric ratio of CO_2 in the oxidizer, i.e., O_2/CO_2 mixture). It is observed that R predicted by OTM increases exponentially with Z. This is because the flame propagation speed decreases dramatically with Z [16] and the radiating time is inversely proportional to flame propagation speed. However, when radiation absorption is considered, R predicted by SNB is shown to be with 1% and thereby negligible. Similar results are shown in Fig. 6(b) for R as a function of equivalence ratio at $Z = 0.6$. Both Fig. 6(a) and (b) indicate that for CO_2-diluted oxy-fuel mixtures, the radiation-induced uncertainty in S_u^0 measurement is exaggerated when radiation absorption is not considered and the uncertainty is in fact negligible since it is greatly reduced by radiation absorption.

Recently, Burke et al. [17] have measured S_u^0 of diluted H_2/O_2 mixtures at high pressures and these data have been popularly used to validate/develop the high-pressure hydrogen mechanism (e.g., [28]). Santner et al. [11] studied the radiation-induced uncertainty of these data. However, they did not consider radiation absorption and thereby the radiation-induced uncertainty was expected to be over-predicted in [11]. This was

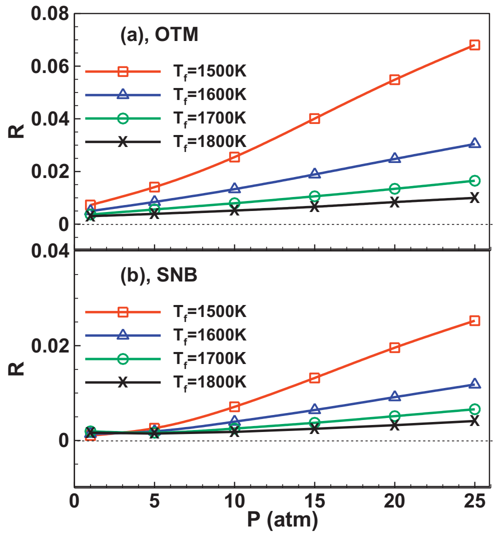

Fig. 7. Relative laminar flame speed reduction, R, as a function of pressure for mixture #6. The results are based on the (a) OTM and (b) SNB models. .

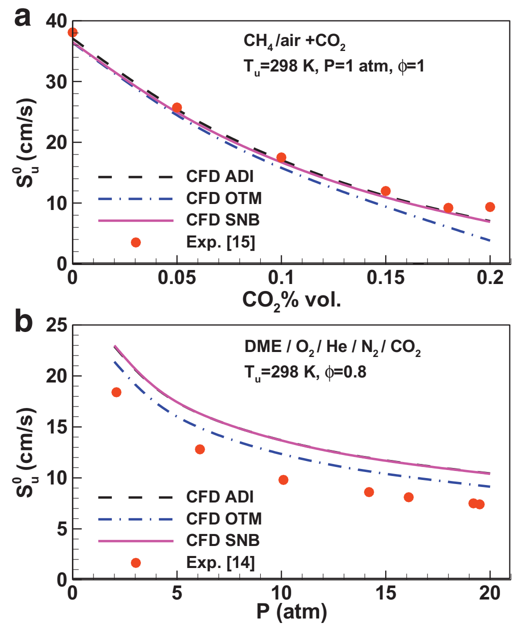

Fig. 9. Comparison of laminar flame speeds measured from experiments and those predicted by simulations using different radiation models: (a), mixture #3 [15]; (b), mixture #2 [14].

confirmed by Fig. 7. Four kinds of $H_2/O_2/CO_2$ mixtures with $\phi = 2.5$ and different adiabatic flame temperatures of $T_f = 1500$–1800 K were considered at a broad range of pressure of $P = 1$–25 atm.

Fig. 7 shows that for both OTM and SNB, R increases as P increases or T_f decreases. This is due to the facts that the radiative flux is nearly proportional to pressure and that the burned gas has longer radiating time at higher P or lower T_f (since the flame speed becomes smaller at higher P or lower T_f). When radiation absorption is not considered as in OTM, R reaches 6.8% at $T_f = 1500$ K and $P = 25$ atm. Such large uncertainty might limit the value of experimental data for kinetic model development and optimization [28]. However, Fig. 7(b) shows that when radiation absorption is considered as in SNB, R is reduced to 2.5% at $T_f = 1500$ K and $P = 25$ atm. Fig. 7(b) indicates that at higher flame temperature of $T_f \geq 1600$ K, R is within 1.2% for $P = 1$–25 atm. Therefore, for mixture #6, the radiation-induced uncertainty in $S_u{}^0$ measurement is also greatly reduced by radiation absorption and it becomes nearly negligible.

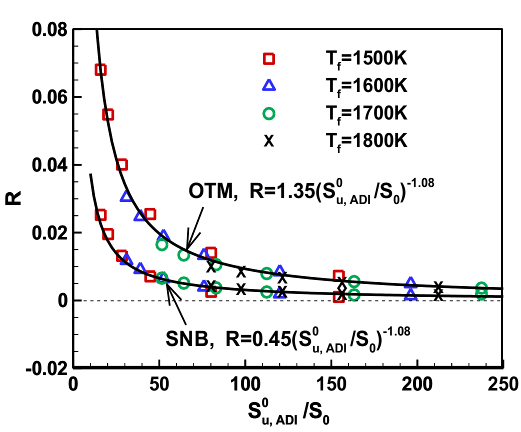

Fig. 8. Relative laminar flame speed reduction, R, as a function of normalized flame speed ($S_0 = 1$ cm/s) for mixture #6.

Fig. 8 plots R as a function of normalized flame speed for different $H_2/O_2/CO_2$ mixtures with $T_f = 1500$–1800 K and $P = 1$–25 atm. All the results (symbols) for OTM and SNB fall almost on two curves, which represent the power-law correlations between R and adiabatic $S_u{}^\circ$ shown in Fig. 8. Moreover, the coefficient in the power-law correlation of SNB, 0.45, is only one-third of that of OTM, 1.35. Such large difference is caused by radiation absorption. Therefore, for mixture #6 the radiation-induced uncertainty in $S_u{}^0$ measurement is also greatly reduced by radiation absorption and it is within 2.5%.

All the results in Figs. 1–8 were obtained from simulations. In Fig. 9 we compared $S_u{}^0$ measured from experiments [14,15] with those from simulations. For mixture #3, Fig. 9(a) shows that compared to experimental data, OTM under predicts $S_u{}^0$, especially at elevated pressures, while SNB well predicts $S_u{}^\circ$ except for case of 20% vol. CO_2, for which the experimental result might be greatly affected by buoyancy. This confirms our conclusions that OTM greatly over-predicts the radiation-induced uncertainty in S_u^0 measurements and that radiation absorption needs to be considered. Fig. 9(b) shows that compared to experimental data, SNB has larger over-prediction than OTM. However, this does not mean that OTM is more accurate than SNB. The disagreement in Fig. 9(b) was mainly due to the fact that the chemical mechanism of DME used in simulation cannot accurately predict S_u^0 at elevated pressures [14].

4. Conclusions

Numerical simulations of spherical flame propagation were conducted for different CO_2 diluted mixtures at a broad range of pressure up to 25 atm. Comparison between results predicted by two radiation models, OTM and SNB, were conducted in order to assess the effects of radiation absorption on spherical flame propagation and radiation-induced uncertainty in laminar flame speed measurement. It was found that for CO_2 diluted mixtures, radiation absorption has great impact on spherical flame propagation and it significantly reduces the radiation-induced thermal and flow effects. Moreover, it was demonstrated that the influence of radiation absorption becomes stronger at higher pressure. The radiation-induced uncertainty in S_u^0 measurement was shown to be substantially over-predicted by OTM and it was greatly reduced by radiation absorption. For all the CO_2 diluted mixtures listed in Table 1, the radiation-induced uncertainty in S_u^0 measurement is almost negligible (within 2.5%) after considering radiation absorption. Therefore, it is expected that for most CO_2 diluted mixtures, the radiation-induced uncertainty in S_u^0 measurement can be neglected.

It is noted that H_2O is another important radiative gas and that it was not considered in the present study. Further investigation needs to be conducted for H_2O diluted mixtures. Nevertheless, it is expected that similar conclusions can be drawn for H_2O diluted and CO_2 diluted mixtures.

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