

Numerical study on chemiluminescence from CH* to estimate total heat release rate in methane-air diffusion flames

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1 Introduction

Chemiluminescence is the spontaneous emission of light from the chemically excited molecules during the transition to their ground states. The most common and strongest chemiluminescence sources in hydrocarbon flames is the excited radicals, such as CH*, OH*, CO₂* and C₂*, where * denotes an excited state, which are usually short-lived during the combustion process. These excited radicals are formed via chemical reactions associated with the combustion event and the chemiluminescence from these radicals is inherently related with the flame parameters. Nevertheless, the measurement of chemiluminescence is relatively simple as compared to the other flame diagnostic tools and non-intrusive nature. Due to these reasons, flame diagnostic concept via measuring chemiluminescence has received a significant attention to understand in-situ time-dependent combustion status in practical combustors. Among the various parameters to be accessed, the heat release rate is considered as one of important parameters not only to characterize strong reactive zone but also to predict and control the unsteady combustion [1, 2]. The direct measurement of temporally and spatially heat release rate in flame is, however, very difficult due to the complexity in applying the currently available measurement techniques to the unsteady combustion environment [3, 4]. So far, numerous studies have been attempted to predict the heat release rate in flames via chemiluminescence numerically as well as experimentally throughout many decades. However, most of the previous attempts were targeted to either premixed or partially premixed combustion [5-9], whereas few studies under limited conditions were reported on non-premixed (diffusion) combustion [10]. The reason of the existence of limited study on diffusion flames might be because the production of soot in typical hydrocarbon fuels' diffusion flames is generally observed and continuous spectrum emission (i.e., thermal radiation) from the soot particles would mask the contribution from the light emissions from excited radicals. However, if it is possible to *separate* the emission from the soot and chemiluminescence via certain way or the target flame to be diagnosed is typical non-sooting diffusion flames (namely, hydrogen-mixed fuel's flame, oxygenated fuel's flame and micro-scale flames whose flame height is less than smoke point), finding correlation between chemiluminescence and heat release in the system should be quite valuable for diagnose such flames. For instance, the optical access to micro-scale diffusion flames is quite difficult especially at near extinction conditions so that chemiluminescence-based diagnostics could play a vital role to study such limiting flames.

Therefore, in this study, we investigate numerically the adequacy of the observable chemiluminescence as the indicator of total heat release rate in 1-D non-sooting counterflow diffusion flames. Methane is chosen as fuel in this study since the combustion reaction mechanism for methane is well established, and also it is widely used in practical combustion systems. The wide range of conditions, such as, velocity, diluents and the chemical reaction models for excited radicals are considered to examine the correlation between total heat release rate and total chemiluminescence due to CH*.

2 Numerical Modelling

2.1 Applied Numerical model: 1-D CFDF

In the present 1-D counterflow configuration, inlet boundaries for fuel and oxidizer stream are spaced 20 mm apart and initial temperature for both fuels and oxidizer flow at the inlets are kept at temperature of 300 K. The methane gas is considered to supply at fuel stream while air is supplied at the oxidizer stream. Fuel gas is diluted by the inert gas species, such as, nitrogen (N₂), water vapor (H₂O), carbon dioxide (CO₂) and Argon (Ar) to the examine their effect on the predicted results. The steady-state 1-D transport equations that governed the counterflow methane-air diffusion flame is solved numerically by Chemkin Software [11] using GRI-Mech 3.0 [12] incorporating with the sub-kinetic model proposed by Nori et al. [13] for chemically-excited radicals OH* and CH* throughout the study unless it is specified. The excited radical, CH* is chosen to examine the total chemiluminescence and total heat release rate correlation as CH* emits light at a wave length of 431 nm which falls in the range that corresponds to visible wavelengths of light. To ensure the generality of the predicted correlation, we also have applied the sub-reaction models for excited radicals proposed by Kojima et al. (2005) [14], Panoutsos et al. (2009) [15], Walsh et al. (1998) [16], and Vries et al. (2007) [17]. Thermodynamics properties for CH* and OH* are taken from the Burcat's thermodynamics data [18] while the transport properties for CH* and OH* are basically considered as the same as their ground state.

2.2 Numerical computation of chemiluminescence

In the present study, our concern is to establish a correlation between the total observable light emission and total heat release rate in non-sooting methane-air counterflow diffusion flames and consequently, CH* is selected due to its light emitting feature as mentioned earlier. Therefore, according to Nori et al. [13], the photon emission rate, I_{CH^*} (mole photons/cm³s) from an excited species CH* is estimated by the relation: $I_{CH^*} = A \times [CH^*]$ where $[CH^*]$ is concentration of CH* and A ($=1.85 \times 10^6 \text{ s}^{-1}$ [13]) is the Einstein coefficient for spontaneous emission for CH* during the transition, i.e. CH* (excited state) \rightarrow CH (ground state), and then the computed CH* photon emission profiles across the flame is integrated over the axial length of L ($= 20 \text{ mm}$) to produce the chemiluminescence emission per unit flame area (mole photons /cm² s) which is given by

$$TCL = \int_0^L I_{CH^*} dx \quad (1)$$

where TCL denotes the total chemiluminescence.

3 Results and discussion

Fig.1 shows the typical flame structure of methane-air diffusion flame without dilution effect for different reaction model for excited radicals when 0.5 m/s ejecting velocity is adjusted at both fuel and oxidizer stream. It is clear that the chemical reaction models for excited radicals don't exhibit any significant influence on the profiles of fuel, oxidizer, heat release and temperature, but the reaction models are found to be significantly sensitive only to the concentration profiles of CH*. Further inspection, it is found that reaction models proposed by Walsh et al. (1998) and Kojima et al. (2005) produce almost the same concentration profile of CH* whereas the reaction models proposed by Nori et al. (2008) and Panoutsos et al. (2009) produce the same concentration profiles of CH* during combustion. It is also seen that the concentration profiles of CH* predicted by the reaction model of Vries et al. (2007) is underestimated as compared to other reaction models considered in this study. However, a slight deviation is found in the peak values and peak location of the concentration profile of CH* between these two groups (Walsh-Kojima and Nori-Panoutsos), and for Vries reaction model, a significant deviation is observed in the peak values of the concentration profile of CH*. This

deviation is supposed to be caused by the production/consumption feature of CH* due to the different reaction paths involved in the reaction models for excited radicals. Besides, a small difference in the peak values of heat release rate is also seen among the adopted reaction models in this study for excited radicals. However, it is interesting to note here that the irrespective of the reaction models for excited radicals, the concentration profile of CH* shows good match with that of the strong positive peak of heat release rate over the narrow zone.

Fig. 2 shows the profiles of major reactions which are accounted for the production and consumption of CH* in methane-air diffusion flames subjected to the same reaction models and conditions applied in Fig.1. It is clear in Fig. 2, for the reaction models proposed by Walsh et al. (1998), Kojima et al. (2005), Nori et al. (2008) and Panoutsos et al. (2009) of excited radicals, that CH* is generated through oxidation reaction of C₂H with O atom and O₂ molecule, respectively (C₂H + O ⇒ CH* + CO; C₂H + O₂ ⇒ CH* + CO₂), while major amount of it is consumed mainly by collision with N₂ (CH* + N₂ ⇒ CH + N₂). Moreover, it is found that the oxidation reaction, C₂H + O ⇒ CH* + CO is found to be play a major role to produce CH* for the reaction models proposed by Walsh et al. (1998) and Kojima et al. (2005) while the reaction, C₂H + O₂ ⇒ CH* + CO₂ plays a negligible role to the production of CH*. On the contrary, the reaction C₂H + O₂ ⇒ CH* + CO₂ is seen to play a little higher role in producing CH* compared to C₂H + O ⇒ CH* + CO for the reaction models proposed by Nori et al. (2008) and Panoutsos et al. (2009). Additionally, it is clear in Fig.2 for the reaction model proposed by Vries et al. (2007) that CH* is generated through the reactions, C₂H + O ⇒ CH* + CO and C₂ + OH ⇒ CH* + CO while major part of CH* is consumed by colliding with H₂O (CH* + H₂O ⇒ CH + H₂O). It is also clear that the oxidation reaction, C₂H + O ⇒ CH* + CO plays a major role to the production of CH* while the reaction, C₂ + OH ⇒ CH* + CO is seen to play a negligible role in producing the excited radical (CH*). Besides, a clear deviation is seen in the peak value and peak position of the major production/consumption rate of CH* for different reaction models and this feature is consistent with the concentration profiles of CH* (Fig.1). Although not shown in figure here, the exactly same trend is found for different reaction models when fuel dilution is employed.

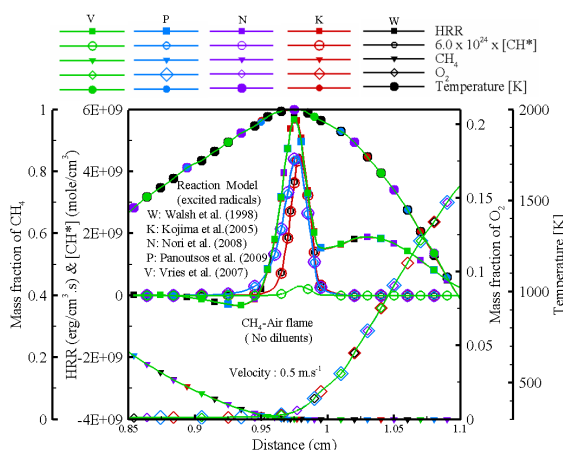


Figure 1. Typical flame structure, heat release rate and concentration of CH*, [CH*] in methane-air diffusion flame under different kinetics model for excited radicals (ejected velocity: 0.5 m/s).

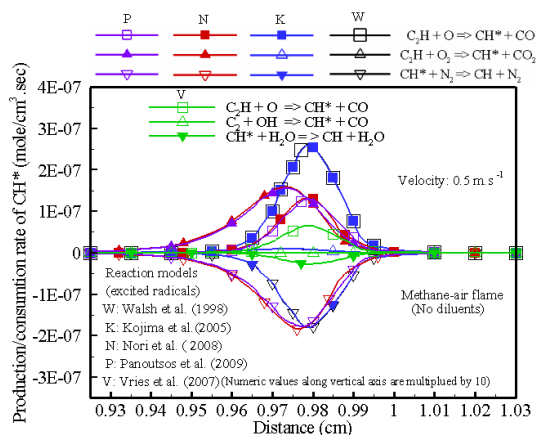


Figure 2. The major reactions for the production and consumption rate of CH* radicals in methane-air diffusion flame under different kinetics model for excited radicals (ejected velocity: 0.5 m/s).

Fig. 3 shows the relation between the total heat release rate and total observable chemiluminescence emission due to CH* in methane-air diffusion flame. It should be mentioned here the total heat release rate is obtained by integrating the profile of heat release rate over the space. Therefore, the total heat release rate and total observable chemiluminescence emission plotted in this figure do not possess the

profile information; rather, these are characteristic values for the imposed conditions. It is seen clearly in figure that they are correlated almost linearly within a moderate range of velocity condition. It is also apparent in Fig. 3 that different kinetics model for excited radicals produce the qualitatively similar feature, although the tangents of the linear-fit-lines are not identical. These non identical

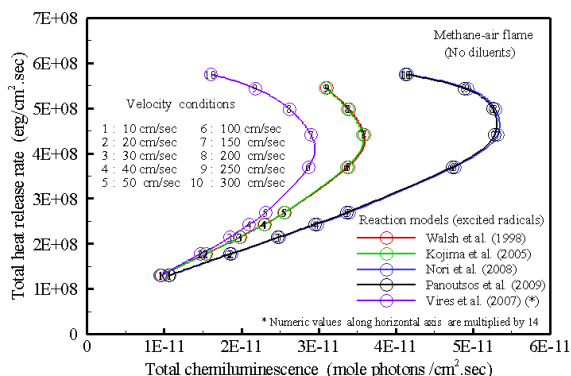


Figure 3. Influence of chemical reaction models for excited radicals on total chemiluminescence and total heat release rate correlation.

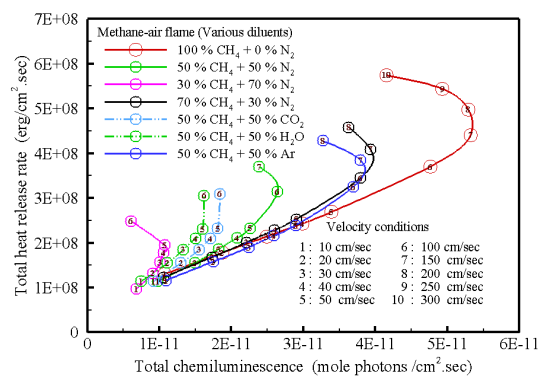


Figure 4. Influence of diluents on total chemiluminescence and total heat release rate correlation.

tangential lines on the linear correlation for different reaction models are connected with the deviation in the peak values of the production/consumption rate of CH* and heat release rate observed in Fig.2 and Fig.1, respectively. As the total observable chemiluminescence emission increases, however, it is noted that the relation becomes slightly away from the linear line, rather slightly bended toward the left in the figure. Such bending trend is also pronounced when the fuel is diluted which is clearly seen Fig. 4. Since the both quantities (i.e. total heat release rate and total observable chemiluminescence emission) increase when the applied velocity increases and approached to the blow-off limit, the observed “bending” might be somehow related to the extinction. We will discuss this issue in next. However, it is worthy to note here that the existence of breaking of the linear relationship for high velocity case is somehow similar to what has been reported by Najm et al. [19], although their target was premixed flame and the breaking source is not the stretch, but the curvature or something else.

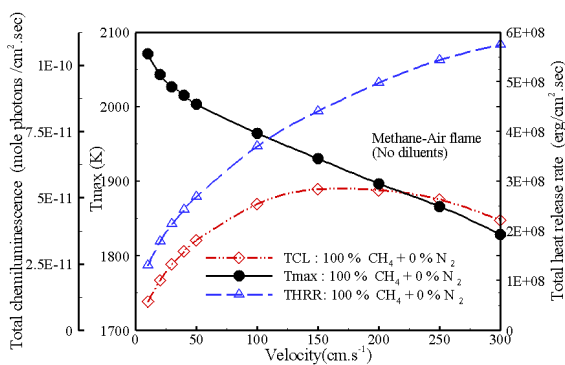


Figure 5. The distribution of maximum temperature (Tmax), total chemiluminescence (TCL) emission and total heat release rate (THRR) against ejecting velocity.

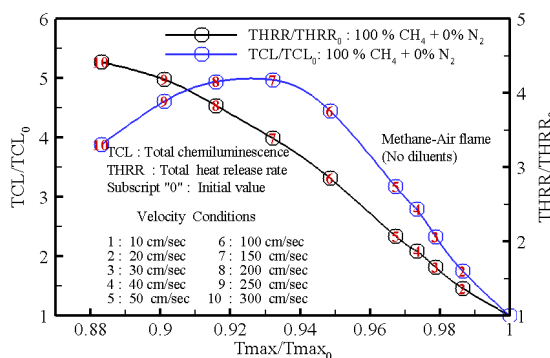


Figure 6. The different temperature sensitivity on total chemiluminescence (normalized) and total heat release rate (normalized).

Fig. 5 represents the response of applied velocity on the distribution of maximum temperature, total chemiluminescence and total heat release rate without dilution effect. It is found in Fig.5 that the decrease in the applied velocity leads to decrease in the maximum temperature. Furthermore, it is seen that the trend of the total chemiluminescence follows the trend of the total heat release rate in lower velocity regime; for higher velocity regime, total chemiluminescence is found to follow the trend of maximum temperature. This feature reflects the fact that the temperature sensitivity is different on these quantities, which is clearly seen in Fig. 6. This different sensitivity of temperature directly causes the non-linear trend in the correlation as described earlier.

The reason why the maximum temperature, not temperature profile, can affect directly on the relationship between total heat release rate and total chemiluminescence might be following two reasons. First, these quantities themselves do not have any spatial information anymore (note that they are calculated by integrating over the space), rather, should be related to the characteristic value, likely maximum temperature, for the prescribed system (\sim condition). Secondary, as described earlier, heat release zone so as to the CH* consumption zone (where chemiluminescence emission is expected) is relatively narrow and the field temperature in the corresponding zone could be characterized by the maximum temperature.

4 Conclusions

In this study, the visible light emission characteristics from a non-sooting methane-air counterflow diffusion flame is investigated numerically, over a wide range of velocity condition and dilution effect, adopting the detailed chemical kinetics of GRI-Mech 3.0 incorporating the chemiluminescence kinetics proposed by Nori et al. (2008). From the present investigation the following conclusions can be drawn.

- (1) The predicted total chemiluminescence emission from methane-air diffusion flame is correlated linearly with total heat release rate for moderate velocity conditions; for high velocity and highly diluted fuel condition, non-linear relationship is observed.
- (2) The main cause of non-linearity is the different sensitivity of temperature on the total heat release rate and total observable chemiluminescence emission.

In the present study, the linear correlation between the total observable light emission and total heat release rate is ensured for the moderate velocity regime and this trend also sustains when different chemical reaction models for excited radicals are applied, which means this linear correlation is the general trend in the moderate velocity regime. Therefore, it is expected that total observable light emission can be utilized to predict the total heat release rate in non-sooting diffusion flame in practical combustors without instrumental complications encountered in using the conventional optical measurement techniques. On the contrary, when the combustion is taken place under very severe condition (with high stretch and local extinction is expected), there is hard to convince that chemiluminescence intensity can tell the total heat release rate in the system.

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