



Critical condition for the ignition of reactant mixture by radical deposition

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Abstract

Motivated by recent interests in plasma assisted combustion (PAC), theoretical investigation on the ignition of pre-mixture by radical deposition is performed. Thermally sensitive intermediate kinetics are considered and the governing equations for temperature and mass fractions of fuel and radical are solved analytically. The correlation depicting the evolution of flame kernel resulted from radical and/or heat deposition is derived. Based on this correlation, we study the flame bifurcation and critical condition for the ignition caused by radical as well as heat deposition. The emphasis is placed on investigating the effects of fuel and radical Lewis numbers. For ignition by radical deposition only, it is demonstrated that the fuel Lewis number has a pronounced influence on the flame propagation and flame bifurcation. With the increase of the fuel Lewis number, new flame branches and bifurcations are observed. It is found that there are two regimes in the change of the minimum chemical ignition power with the fuel Lewis number. Unlike the fuel Lewis number, the radical Lewis number only has a quantitative influence on the flame bifurcation and critical ignition condition. For ignition by radical and thermal deposition, the flame trajectory and flame bifurcation are found to be strongly affected by additional heat deposition. The minimum chemical and thermal ignition powers are compared, and the ignition efficiency of pure chemical ignition and pure thermal ignition is shown to strongly depend on the fuel and radical Lewis numbers. Moreover, the validity of theoretical results describing the effects of fuel and radical Lewis numbers on the minimum ignition energy is confirmed qualitatively by transient numerical simulations including thermal expansion and detailed chemistry.

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1. Introduction

Ignition is the process whereby a medium capable of reacting exothermically is brought to a state of rapid combustion and it is one of the most important problems in combustion. Understanding of the ignition process and critical ignition condition is important not only for fundamental combustion research but also for developing high

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performance combustion engines. It is well known that successful ignition depends on the amount of energy in the form of heat and/or radicals deposited into a combustible mixture. If the energy is smaller than the so-called minimum ignition energy (MIE), the resulting flame kernel decays rapidly because heat/radicals conducts/diffuse away from the kernel and the dissociated species recombine faster than they are generated by chemical reactions within the ignition kernel [1].

In the literature, there are many theoretical studies on the ignition process [1–9]. However, all these studies only consider ignition caused by heat deposition, and there is no theoretical study on the ignition by radical deposition. In practical ignition process, radicals as well as heat are generated by the spark and these radicals also support flame kernel propagation. In fact, in recent studies on plasma assisted combustion (PAC) [10–12], the chemical effect of radical generated by non-equilibrium plasma is found to be much stronger than the thermal effect on ignition and flame stabilization. Therefore, it is of great significance to understand the ignition by radical deposition.

Some numerical studies on the ignition by radical have been conducted. For examples, Dixon-Lewis and Shepherd [13] studied the influence of radicals appeared in the initial ignition stage on the ignition process, Wiriyawit and Dabora [14] investigated the combined effects of chemical and thermal ignition in hydrogen/air mixtures, Sloane et al. [15,16] studied the influence of chemical and thermal ignition on the MIE and ignition delay, and Beduneau et al. [17] examined the effects of radicals generated in different stages of spark ignition on the flame kernel evolution. However, since chemical ignition and thermal ignition exist simultaneously in all these studies [13–17], an individual investigation on chemical ignition could not be performed. Furthermore, numerical simulation is limited to specific fuel/air mixture, and the effects of Lewis number remain unclear. Therefore, to gain a general understanding of chemical ignition, it is necessary to perform an analytical study.

The objectives of this study are to theoretically analyze the ignition by radical deposition and to examine the critical ignition condition. In the following, the mathematical model and theoretical analysis are first presented. Then, results and discussions on flame bifurcation and critical ignition condition are shown in Section 3. In order to validate theoretical results, detailed numerical simulations are conducted in Section 4. Finally, the conclusions are presented in Section 5.

2. Theoretical analysis

In order to study the ignition by radical deposition, chain-branching kinetics of intermediate species (radicals) should be considered. Similar

to our previous work on ignition by heat deposition [9], we employ the simplified Zel'dovich-Liñán model proposed by Dold and coworkers [18,19]. This model comprises a chain branching reaction $F + Z \rightarrow 2Z$, and a recombination reaction $Z + M \rightarrow P + M$, where F , Z , and P represent fuel, radical, and product, respectively, and M denotes any type of molecule. The branching reaction is thermally sensitive and has a rate constant depending on the temperature in the Arrhenius form, while the rate of the recombination reaction is independent of the temperature [18,19]. This model was used in previous studies on the ignition, propagation, extinction, and stability of premixed flames [9,18–21].

One-dimensional spherical flame initiation is considered and the mathematical model is similar to that in Ref. [9]. The constant density and quasi-steady assumptions [6–9] are employed. In the coordinate attached to the propagating flame front and under the assumption of large activation energy, the non-dimensional governing equations for temperature, T , and mass fractions of fuel, Y_F , and radical, Y_Z , in the unburned and burned zones are [9]

$$-U \frac{dY_F}{dr} = \frac{1}{Le_F} \frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{dY_F}{dr} \right) \quad (1a)$$

$$-U \frac{dY_Z}{dr} = \frac{1}{Le_Z} \frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{dY_Z}{dr} \right) - Y_Z \quad (1b)$$

$$-U \frac{dT}{dr} = \frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{dT}{dr} \right) + QY_Z \quad (1c)$$

where r is the non-dimensional radial coordinate. Le_F and Le_Z are the Lewis numbers of the fuel and radical, respectively. U is propagation speed of the flame front, and Q the specific heat release of the recombination reaction.

Spherical flame initiation can be achieved by heat and/or radical deposition. In the quasi-steady model, the ignition energy is provided as a heat or radical flux at the center. Therefore, the boundary conditions are

$$r \rightarrow 0 : r^2 \frac{dY_F}{dr} = 0, \quad r^2 \frac{dY_Z}{dr} = -\frac{Le_Z}{Q} q_c, \quad r^2 \frac{dT}{dr} = -q_t \quad (2a)$$

$$r \rightarrow \infty : Y_F = 1, Y_Z = 0, T = 0 \quad (2b)$$

where q_c and q_t are referred to as the chemical and thermal ignition powers, respectively. They are equal to the dimensional ignition power normalized by the same scaling quantity, $4\pi r_s^2 \lambda T_s$, and thus can be directly compared. Physically, q_t is the non-dimensional thermal flux while q_c is the enthalpy flux corresponding to the mass diffusion of radical.

The jump conditions across or at the flame front ($r = R$) [18,19] are

$$[Y_F] = [Y_Z] = [T] = \left[\frac{dT}{dr} \right] = \left[\frac{1}{Le_F} \frac{dY_F}{dr} + \frac{1}{Le_Z} \frac{dY_Z}{dr} \right] = T - 1 = Y_F \frac{dT}{dr} = 0 \quad (3)$$

The analytical solutions to Eqs. (1–3) are directly presented here with the detailed derivations shown in the **Supplementary data**. The distribution for the fuel mass fraction is obtained as

$$Y_F(r) = \begin{cases} 0 & \text{for } 0 \leq r \leq R \\ 1 - \int_r^\infty \xi^{-2} e^{-Le_F U \xi} d\xi / \int_R^\infty \xi^{-2} e^{-Le_F U \xi} d\xi & \text{for } r \geq R \end{cases} \quad (4)$$

The solution for the radical mass fraction is

$$Y_Z(r) = \begin{cases} Y_{Zf} e^{0.5(ULe_Z+k)(R-r)} \frac{F(kr, ULe_Z/k, -ULe_Z/k)}{F(kR, ULe_Z/k, -ULe_Z/k)} - \frac{Le_Z}{Q} q_c \cdot \vartheta(r) & \text{for } 0 \leq r \leq R \\ Y_{Zf} e^{0.5(ULe_Z+k)(R-r)} \frac{G(-kr, ULe_Z/k, -ULe_Z/k)}{G(-kR, ULe_Z/k, -ULe_Z/k)} & \text{for } r \geq R \end{cases} \quad (5)$$

in which $F(a, b, c) = \int_0^1 e^{at} t^b (1-t)^c dt$, $G(a, b, c) = \int_0^\infty e^{at} t^b (1+t)^c dt$, $k = \sqrt{(ULE_Z)^2 + 4Le_Z}$, and

$$\vartheta(r) = k \cdot e^{-0.5r(ULe_Z+k)} \left[\frac{G(-kR, ULe_Z/k, -ULe_Z/k) F(kr, ULe_Z/k, -ULe_Z/k)}{F(kR, ULe_Z/k, -ULe_Z/k)} - G(-kr, ULe_Z/k, -ULe_Z/k) \right] \quad (6)$$

In Eq. (5), the radical mass fraction at the flame front, Y_{Zf} , is determined according to the jump condition $[Le_F^{-1} dY_F/dr + Le_Z^{-1} dY_Z/dr = 0]$ at $r = R$ in Eq. (3) and has the following form

$$Y_{Zf} = \frac{Le_Z Le_F^{-1} k^{-1} R^{-2} e^{-Le_F UR} / \int_R^\infty \xi^{-2} e^{-Le_F U \xi} d\xi}{\frac{F(kR, 1+ULe_Z/k, -ULe_Z/k)}{F(kR, ULe_Z/k, -ULe_Z/k)} + \frac{G(-kR, 1+ULe_Z/k, -ULe_Z/k)}{G(-kR, ULe_Z/k, -ULe_Z/k)}} + Le_Z Q^{-1} q_c k e^{-\frac{1}{2}R(ULe_Z+k)} G(-kR, ULe_Z/k, -ULe_Z/k) \quad (7)$$

The temperature distribution is

$$T(r) = \begin{cases} 1 + \int_0^r \int_0^s I(s, \xi) d\xi ds + q_t \int_r^\infty s^{-2} e^{-Us} ds & \text{for } 0 \leq r \leq R \\ [1 + \int_r^\infty \int_s^\infty I(s, \xi) d\xi ds] \frac{\int_r^\infty s^{-2} e^{-Us} ds}{\int_R^\infty s^{-2} e^{-Us} ds} - \int_r^\infty \int_s^\infty I(s, \xi) d\xi ds & \text{for } r \geq R \end{cases} \quad (8)$$

where $I(s, \xi) = (\xi/s)^2 e^{-U(s-\xi)} Q Y_Z(\xi)$.

Substituting the above temperature distribution into the condition $[dT/dr] = 0$ at $r = R$ in Eq. (3), we obtain the following correlation depicting the change of the flame propagation speed U with the flame radius R

$$\int_R^\infty \int_0^s I(s, \xi) d\xi ds + q_t \int_r^\infty s^{-2} e^{-Us} ds = 1 \quad (9)$$

which includes chemical and thermal ignition powers (q_c and q_t), Lewis numbers of fuel and radical (Le_F and Le_Z), and heat release (Q).

By numerically solving Eq. (9), the flame bifurcations and regimes at different Lewis numbers and/or ignition powers can be obtained, and thereby the critical ignition condition and mini-

mum ignition power can be determined. Since this study is mainly focused on the effects of fuel and radical Lewis numbers on ignition, in the following we present results obtained from Eq. (9) at different values of (Le_F, Le_Z) and the non-dimensional heat release is fixed to be $Q = 2.0$, a value that corresponds to a typical hydrocarbon mixture with initial temperature at 300–500 K [19].

3. Results and discussion

3.1. Ignition by radical deposition only

We first consider ignition caused by radical deposition only and thereby the heat flux from the center is set to be zero ($q_t = 0$). Figure 1 shows the change of the flame propagation speed with the flame radius for $Le_F = Le_Z = 1.0$. The intersection points of the curves with the horizontal axis in Fig. 1 represent flame ball solutions ($U \rightarrow 0$). It is seen that at $q_c = 0$, the spherical flame can propagate outwardly only when its radius is larger than the flame ball radius $R_Z \approx 0.8$. At $q_c = 0.4$ or 0.8 , the $U-R$ curve is shown to be shifted toward the left side and thus the flame ball radius becomes smaller. However successful ignition is not achieved. By further increasing q_c to 1.0, the $U-R$ curve critically crosses the origin point at $R = U = 0$ (see the inset in Fig. 1), indicating that the spherical flame kernel can successfully propagate outwardly along the $U-R$ curve to eventually become a planar flame ($R \rightarrow \infty$). Therefore, the minimum chemical ignition power for $Le_F = Le_Z = 1.0$ is $q_{c,min} = 1.0$. When $q_c = 1.2 > q_{c,min}$, Fig. 1 shows that the flame propagation is promoted at small flame radius. At large flame radius ($R > 5$), the radical ignition is shown to have no influence on the flame propaga-

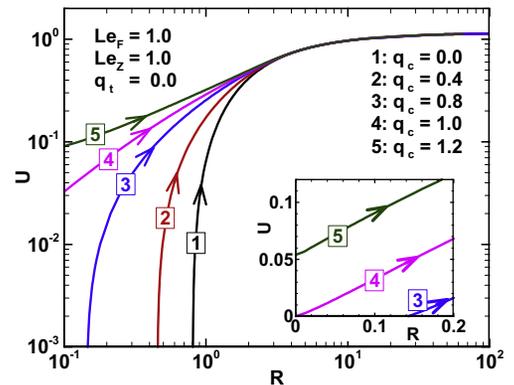


Fig. 1. Flame propagation speed as a function of flame radius at different chemical ignition powers.

tion speed. This is consistent with numerical and experimental results in Refs. [22,23].

To assess fuel Lewis number effects, some typical results are presented in Fig. 2, which shows different flame bifurcations from those for $Le_F < 2.0$. For $Le_F = 2.5$, Fig. 2(a) shows that at zero and low chemical ignition powers ($q_c = 0, 0.4$ and 0.8), the $U-R$ curves are C-shaped and there is no flame ball solution. When q_c is increased to 0.94 or 0.98 , besides the C-shaped flame branch, a new bell-shaped flame branch (with two flame ball solutions) appears. At $q_c = 1.0$, the bell-shaped flame branch starts from the origin point $U = R = 0$, which is similar to results in Fig. 1. However, the spherical flame cannot be successfully initiated at $q_c = 1.0$ since the flame extinguishes with finite flame propagation speed at $R \approx 2.7$ along the bell-shaped $U-R$ curve. This extinguishment is caused by the fact that less radical (due to larger flame radius) and less fuel (due to relatively large Le_F) can diffuse into the reaction zone from the center and thereby the flame cannot propagate outwardly in a self-sustained manner. Further increasing q_c to 1.06 , flame bifurcation occurs: the C-shaped and bell-shaped curves merge and generate the upper

stable flame propagation branch and the lower unstable one. A spherical flame can thereby propagate outwardly along the upper branch and successful ignition can be achieved now. Therefore, we have $q_{c,min} = 1.06$ for $Le_F = 2.5$. Figure 2(b) presents the results for a much larger fuel Lewis number of $Le_F = 4.0$. When q_c is less than unity ($q_c = 0$ and 0.8), only the C-shaped $U-R$ curve exists. As the ignition power is increased to be above unity ($q_c = 1.2$ and 1.6), a new left flame kernel branch occurs. However, the ignition power is still not high enough to achieve successful ignition. Only when the minimum chemical ignition power, $q_{c,min} = 3.288$, is reached, does the flame bifurcation occur and is the flame successfully initialized.

In above results, the radical Lewis number is fixed to be unity ($Le_Z = 1$). Actually, it is found from the results not presented here that Le_Z only has a quantitative influence on the ignition by radical deposition. According to results shown in Figs. 1 and 2, we find that $q_c = 1.0$ is critical for ignition by radical deposition and that both flame bifurcation and critical ignition condition change significantly with the fuel Lewis number. To interpret these observations, we derive the following correlation (which implicitly describes the change of the flame ball radius, R_Z , with the ignition powers, q_c and q_t) from Eq. (9) in the limit of $U = 0$:

$$q_c \sqrt{Le_Z} = \frac{R_Z \sqrt{Le_Z} - Q Le_F^{-1} (R_Z \sqrt{Le_Z} + [\exp(-2R_Z \sqrt{Le_Z}) - 1]/2) - q_t \sqrt{Le_Z}}{1 - \exp(-R_Z \sqrt{Le_Z})} \quad (10)$$

Using Eq. (10), R_Z is plotted as a function of q_c at $q_t = 0$ in Fig. 3. According to Eq. (10), $\sqrt{Le_Z}$ can be scaled into R_Z and q_c (see Fig. 3), which explains why Le_Z does not qualitatively affect the flame bifurcation and critical ignition condition. Figure 3 shows that for $Le_F < 2.0$, R_Z monotonically decreases with q_c and there always exists one flame ball solution for $0 \leq q_c Le_Z^{0.5} \leq 1.0$. For $Le_F = 2.0$, there is no flame ball solution at $0 \leq q_c Le_Z^{0.5} \leq 0.5$ and finite value of R_Z exists only for $0.5 < q_c Le_Z^{0.5} < 1.0$. For $Le_F = 2.5$, there are two flame ball solutions for $0.86 \leq q_c Le_Z^{0.5} \leq 1.0$, which correspond to the two intersection points of the bell-shaped curve with the horizontal axis shown in Fig. 2(a). At $q_c Le_Z^{0.5} > 1.0$, there is only one flame ball solution and R_Z increases monotonically with q_c . For $Le_F = 4.0$, the only flame ball solution exists when $q_c Le_Z^{0.5} \geq 1.0$, which is consistent with the results shown in Fig. 2(b). Therefore, the change of the number of flame ball solutions determines the flame bifurcation. Furthermore, Fig. 3 as well as Eq. (10) demonstrates that all the curves converge to $R_Z = 0$ and $q_c Le_Z^{0.5} = 1$. This explains why at $Le_Z = 1$ the solution of $U = R = 0$ always exists for $q_c = 1$, as shown in Figs. 1 and 2. Besides, from Figs. 1–3 it is seen that for Le_F below some critical value

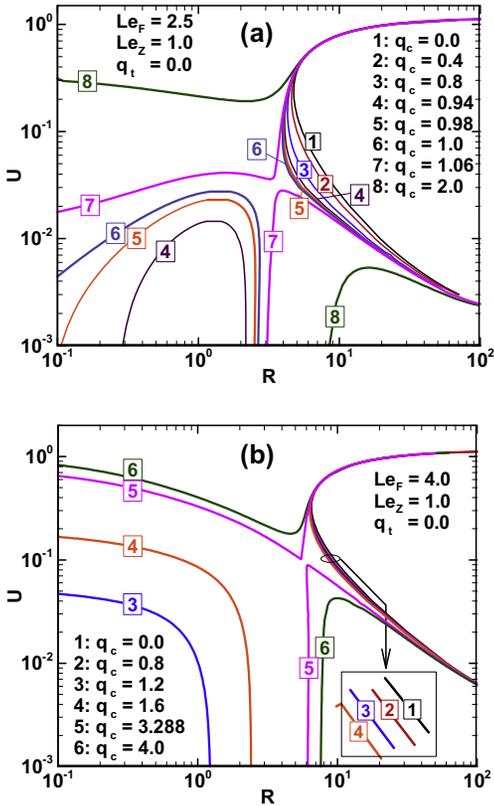


Fig. 2. Flame propagating speed as a function of flame radius for (a) $Le_F = 2.5$; (b) $Le_F = 4.0$.

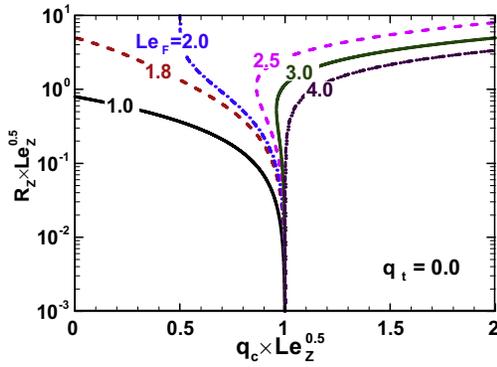


Fig. 3. Change of the flame ball radius with the chemical ignition power for different fuel Lewis numbers.

around 2.5, the critical ignition condition corresponds to $R_Z \rightarrow 0$, while for larger Le_F , the critical ignition occurs when the left and right $U-R$ branches merge (flame bifurcation happens). The latter is also observed in our previous work on ignition by heat deposition [9].

According to the above analysis, the minimum chemical ignition power, $q_{c,min}$, can be obtained for different fuel and/or radical Lewis numbers. The results are summarized in Fig. 4. When the radical Lewis number is fixed, Fig. 4(a) shows that there are two regimes in the change of $q_{c,min}$ with Le_F . In the first regime with $Le_F < Le_F^*$ (~ 2.5), $q_{c,min} = Le_Z^{-0.5}$ is a constant; while in the second regime with $Le_F > Le_F^*$, $q_{c,min}$ increases monotonically with Le_F . The constant value of $q_{c,min} = Le_Z^{-0.5}$ in the first regime is caused by the fact that for $q_c = Le_Z^{-0.5}$, there is always a $U-R$ curve from $R = 0$ to $R \rightarrow \infty$ along which successful ignition can be achieved (see Figs. 1 and 3). The increase of the $q_{c,min}$ with Le_F in the second regime is mainly due to the coupling between the positive stretch rate and the preferential diffusion between heat and fuel (i.e. Le_F) [8,9].

Compared to the influence of Le_F , Fig. 4(b) shows that the influence of Le_Z on $q_{c,min}$ is less pronounced. It is seen that $q_{c,min}$ monotonically decreases with Le_Z . This is due to the fact that weaker mass diffusivity of radicals (thus larger Le_Z) can render them less inclined to diffuse away from the ignition kernel and consequently the radical accumulation induces explosive chain branching reaction which promotes ignition. Besides, since the Markstein length decreases with Le_Z [9] and the flame kernel is highly positively stretched, the spherical flame can be more easily ignited for larger Le_Z .

3.2. Ignition by radical and heat deposition

In the above subsection and our previous work [9], ignition by radical deposition only and heat

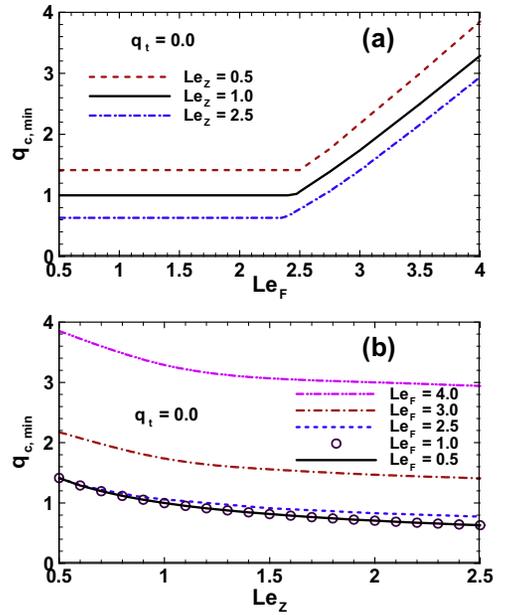


Fig. 4. Change of the minimum chemical ignition power with (a) fuel and (b) radical Lewis numbers.

deposition only are investigated, respectively. However, in practical ignition process, radical and heat deposition occurs simultaneously. In the following, ignition caused by heat and radical is studied based on Eq. (9).

Figure 5 presents the results at different chemical ignition powers but fixed thermal ignition power, $q_t = 0.05$. For $Le_F = Le_Z = 1.0$, comparison between Figs. 5(a) and 1 indicates that there is significant change in the flame trajectory ($U-R$ curves) caused by additional heat deposition. A new flame kernel branch starting from $R = 0$ appears when q_c is below the critical value. When $q_c > q_{c,min} = 0.448$, the left and right flame branches merge with each other and successful ignition can be achieved. Unlike the pure radical ignition shown in Fig. 1 for which the critical ignition happens when the flame trajectory starts from $U = R = 0$, here the critical ignition is reached when the two ball solutions on these two branches becomes the same.

To examine the fuel Lewis number effects, the flame trajectories for $Le_F = 2.2$ is shown in Fig. 5(b). It is seen that for $q_c < 0.67$, there are two flame branches: the left flame kernel one and the right C-shaped one, and there is only one flame ball solution. When q_c is increased to 0.675, a new bell-shaped branch appears in the middle and there are three flame ball solutions now. Further increasing q_c to 0.7 makes the bell- and C-shaped branches merge and the first flame bifurcation occurs, which is similar to the pure radical ignition shown in Fig. 2(a). However,

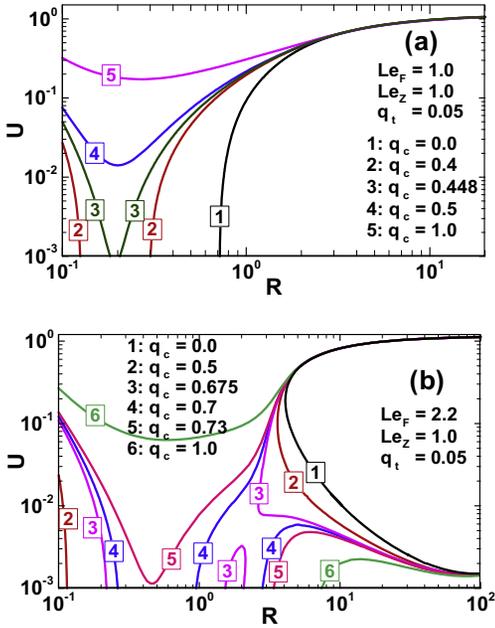


Fig. 5. Flame propagation speed as a function of flame radius at $q_t = 0.05$ for (a) $Le_F = 1.0$; and (b) $Le_F = 2.2$.

successful ignition is still not achieved. Figure 5(b) shows that at $q_c = 0.73$, the left and bell-shaped branches merge and the second bifurcation happens. Now the spherical flame kernel can successfully propagate outwardly. It is observed that the number of the flame ball solutions is reduced from three to one when the critical ignition condition is reached.

Similar to Fig. 3 for pure radical ignition ($q_t = 0$), Fig. 6 demonstrates the effects of Le_F and Le_Z on R_Z when both radical deposition and heat deposition exist. The flame bifurcations and critical ignition conditions shown in Fig. 5 can be explained by Fig. 6. For $Le_F < 2.0$ (the case shown in Fig. 5(a)), there are two flame ball solutions when $q_c < q_{c,min}$. The turning point corresponds to the critical ignition condition at which the two ball solutions disappear. For $2.0 < Le_F < 2.43$, Fig. 6 shows that the C-shaped curve becomes S-shaped and there are two turning points. According to the discussions on Fig. 5(b), the critical ignition condition (the second flame bifurcation) corresponds to the right turning point of the S-shaped curve. Increasing Le_F to 2.5, 3.0 or 4.0, no turning point appears and the only flame ball solution always exists. Therefore, at large fuel Lewis number, the critical ignition condition cannot be obtained according to flame ball solutions. The same conclusion was drawn for ignition by heat deposition only [8,9].

Comparison between Figs. 5(a) and 1 also indicates that the minimum ignition power is reduced roughly by 50% when there are both heat and

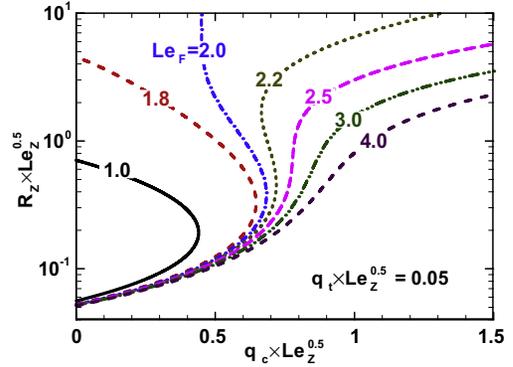


Fig. 6. Change of the flame ball radius with the chemical ignition power for different fuel Lewis numbers.

radical deposition. The critical power for ignition caused by heat and radical deposition are studied and the results are shown in Fig. 7. In Fig. 7, successful ignition occurs only when the ignition power (q_t, q_c) is on the upper right side of the curve. It is seen that at fixed value of chemical (thermal) ignition power, the critical thermal (chemical) ignition power for successful ignition increases with Le_F while decreases with Le_Z . This is consistent with results from pure radical-induced ignition shown in Fig. 4 and results from pure heat-induced ignition in Ref. [9]. Figures 7(a) and (b) indicates that Le_Z only quantitatively affects the critical ignition powers. However, Fig. 7(c) shows that the critical ignition curve is greatly shifted to the right side when the Le_F increases. For pure radical ignition ($q_t = 0$), the critical chemical ignition energy, q_c , is only slightly affected by Le_F while the for pure thermal ignition ($q_c = 0$), the critical q_t increases significantly with Le_F . This is also consistent with results shown in Fig. 4 and Ref. [9].

Furthermore, Fig. 7 also indicates that the critical thermal ignition power is usually lower than the critical chemical ignition power. The comparison between $q_{c,min}$ and $q_{t,min}$ for pure chemical ignition ($q_t = 0$) and pure thermal ignition ($q_c = 0$), respectively, is shown in Fig. 8. It is demonstrated that for most cases, heat deposition is more efficient than radical addition (i.e. $q_{c,min} > q_{t,min}$), and that $q_{c,min} < q_{t,min}$ happens only for mixtures with large Le_F and small Le_Z . Due to the facts that the rate of the branching reaction depends on temperature in the Arrhenius form and that the rate of the recombination reaction is a constant independent of the temperature, the enhancement on the reactivity by heat deposition (which results in very high temperature near the center) is much stronger than that by radical addition. Consequently, ignition by heat deposition is more easily achieved than by radical deposition (i.e. $q_{c,min} > q_{t,min}$). However, for

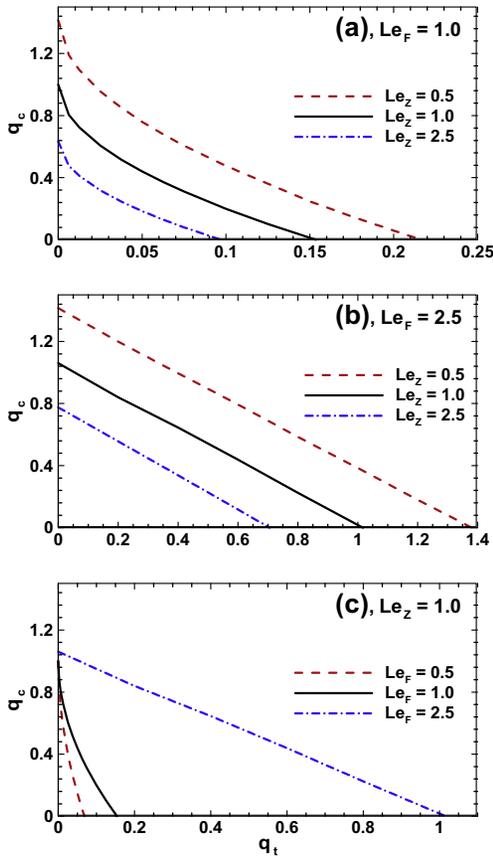


Fig. 7. The critical ignition power for ignition caused by heat and radical deposition for (a) $Le_F = 1.0$; (b) $Le_F = 2.5$; and (c) $Le_Z = 1.0$.

mixtures with large Le_F and small Le_Z (the gray region in Fig. 8), the fuel has small mass diffusivity and thus cannot be effectively transported to the flame front, while the radical can quickly diffuse

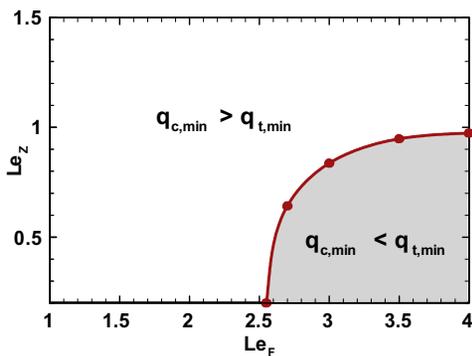


Fig. 8. The comparison between the minimum chemical and thermal ignition powers.

to the flame front from radical source at the center. Therefore, in this case, ignition by radical deposition is more easily achieved than by heat addition (i.e. $q_{c,min} < q_{t,min}$).

It is noted that homogenous ignition is mainly controlled by chemical kinetics so that the ignition delay can be significantly reduced by radical and activated species generated by plasma in PAC. However, in the flame initiation process, the flame kernel is highly stretched and the ignition is mainly controlled by transport properties [8]. Therefore, the critical ignition energy cannot be drastically reduced in PAC.

4. Numerical validation

The above theoretical analysis is based on some assumptions (e.g. constant density, quasi-steady flame propagation, and large activation energy). To qualitatively validate the above theoretical results, transient numerical simulations are conducted by using the in-house code A-SURF to simulate the ignition caused by radical deposition. A-SURF solves the conservation equations of one-dimensional, compressible, multi-component, reactive flow in a spherical coordinate using the finite volume method and it has been successfully used in our previous studies [8,22,24–26]. The details on the governing equations, numerical schemes, and code validation can be found in Refs. [22,24] and thus are not repeated here.

We conduct simulations for premixed $H_2/O_2/He$ mixture with the mole ratio of $H_2:O_2:He = 1:1:8$ at normal temperature and pressure (300 K, 1 atm). The detailed mechanism for hydrogen oxidation [27] is employed. The computational domain is $0 \leq r \leq 50$ cm and a multi-level, dynamically adaptive mesh with the finest mesh size of $8 \mu m$ is used. Different from the theoretical analysis, radical deposition as an initial condition is used in the simulation. The initial distribution of the mass fraction of H radical is assumed to be Gaussian and its full width at half maximum (FWHM) is 1.6 mm. The chemical ignition energy is calculated by integrating the formation enthalpy of H radical over the whole computational domain. The peak mass fraction of H radical at the center is modified to obtain the minimum ignition energy using the method of trial-and-error with relative error below 1%. To examine the influence of fuel and radical Lewis numbers, the mass diffusivities of H_2 and H are artificially changed in the simulation [9]. For example, the radical Lewis number is modified to half of the original values (denoted by the superscript 0), i.e. $Le_H = 0.5Le_H^0$, through changing the mass diffusivity of H from D_H to $2D_H$. Based on the thermodynamic and transport data at 298 K, we have $Le_{H_2}^0 = 0.902$ and $Le_H^0 = 0.587$.

Figure 9 presents the change of minimum chemical ignition energy with the fuel and radical

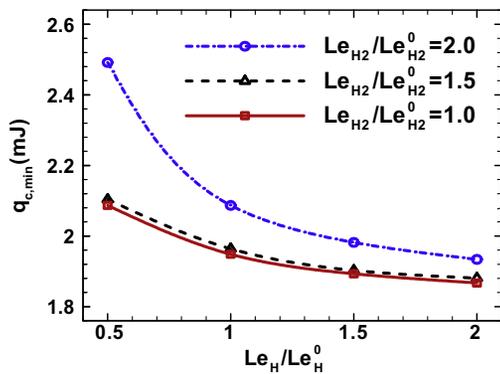


Fig. 9. Effects of fuel and radical Lewis numbers on the minimum chemical ignition energy of $H_2/O_2/He$ mixture.

Lewis numbers. It is seen that in each curve corresponding to the fixed fuel Lewis number ($Le_{H_2}/Le_{H_2}^0 = 1.0, 1.5, \text{ or } 2.0$), the minimum chemical ignition energy, $q_{c,min}$, monotonically decreases with the radical Lewis number, Le_H/Le_H^0 . This agrees qualitatively with the theoretical results in Fig. 4(b). At fixed radical Lewis number (e.g. $Le_H/Le_H^0 = 1.0$), $q_{c,min}$ is shown to increase with Le_{H_2} . Besides, $q_{c,min}$ for $Le_{H_2}/Le_{H_2}^0 = 1.0$ is very close to that for $Le_{H_2}/Le_{H_2}^0 = 1.5$, indicating that the fuel Lewis number does not have pronounced effects on $q_{c,min}$ when it is relatively small. However, $q_{c,min}$ for $Le_{H_2}/Le_{H_2}^0 = 2.0$ is shown to be much larger than that for $Le_{H_2}/Le_{H_2}^0 = 1.5$. Again these observations qualitatively confirm the theoretical results in Fig. 4(b). Therefore, the detailed numerical simulations qualitatively validate the results from theoretical analysis.

5. Conclusions

Ignition by radical and/or heat deposition is studied theoretically using the simplified Zel'dovich-Liñán model proposed by Dold and coworkers [18,19]. The correlation describing spherical flame propagation is derived, based on which flame bifurcation and critical ignition condition are investigated.

For ignition by radical deposition only, the flame bifurcation and critical ignition condition are strongly affected by the fuel Lewis number while the radical Lewis number only has a quantitative influence. The change of the number of flame ball solutions with the chemical ignition power is found to determine the flame bifurcation and critical ignition condition. Two regimes are observed in the change of the minimum chemical ignition power with the fuel Lewis number. With the increase of the fuel/radical Lewis number,

the minimum chemical ignition power is shown to monotonically increase/decrease.

For ignition by radical and heat deposition, great change in the flame trajectory and flame bifurcation is caused by additional heat deposition. Moreover, compared to pure radical ignition, the minimum ignition power is shown to be greatly reduced. The minimum chemical and thermal ignition powers are compared and it is found that for most cases, heat deposition is more efficient in ignition than radical addition. Only for mixtures with large fuel Lewis number and small radical Lewis number is the chemical ignition more efficient than the thermal ignition.

Transient numerical simulations on the ignition by radical deposition for $H_2/O_2/He$ mixture are conducted. The results qualitatively confirm the effects of fuel and radical Lewis numbers on ignition observed in theoretical analysis.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.proci.2012.06.035>.

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