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Effects of hydrogen addition on non-premixed ignition of iso-octane by hot air in a diffusion layer



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ABSTRACT

Hydrogen addition is widely used to improve the combustion performance of single-component fuel. In this study, the effects of hydrogen addition on non-premixed ignition of iso-octane by hot air in a diffusion layer were examined and interpreted numerically. Detailed chemistry and transport were considered in simulation. The non-premixed ignition delay times at different hydrogen blending levels were obtained and analyzed. It was found that hydrogen addition greatly reduces the ignition delay. This is mainly due to the fact that the preferential mass diffusion of hydrogen over iso-octane significantly increases the local hydrogen blending level at the ignition kernel. Besides, for the non-premixed ignition process, two modes of reaction front propagation were identified through the analysis based on Damköhler number and consumption speeds. One is the reaction-driven mode characterized by local or sequential homogeneous autoignition; and the other is the diffusion-driven mode, which depends on the balance of mass diffusion, heat transfer and chemical reaction. These two modes lead to different ignition behaviors. For pure iso-octane with low mass diffusivity, ignition is mainly caused by local homogeneous reaction occurring at the most reactive position. With the increase of diffusion layer thickness, the local temperature at the most reactive position increases and therefore the non-premixed ignition delay time of pure iso-octane decreases. However, when hydrogen with high mass diffusivity is added into iso-octane, the non-premixed ignition is controlled by fuel diffusion. With the increase of diffusion layer thickness, the concentration gradient becomes smaller and thereby less hydrogen diffuses into the ignition kernel. Consequently, unlike pure iso-octane, the non-premixed ignition delay time of hydrogen/iso-octane blends increases with the diffusion layer thickness.

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turbulence intensity. They observed that the incipient ignition spot

is always associated with the most reactive fraction and it is

1. Introduction

Non-premixed ignition and combustion in a diffusion layer happen in various combustion systems. For example, the configuration of fuel-jet diffusion flame is widely used in industry furnaces [1,2] and propulsion systems [3]. For non-premixed ignition and combustion in these systems, one of the important parameters is the ignition delay time, which affects the lift-off distance between the flame and injector, flame stabilization and operation modes of the combustor [4–6].

Fundamental studies on the ignition of laminar diffusion flame have been widely conducted in the literature (e.g., [7–19]). Mastorakos et al. [7,13] and Hilbert et al. [8] investigated the ignition process in a planar diffusion layer under a certain range of nearly independent of turbulence intensity. Similar observations were made in counterflow diffusion flames [16-18]. Besides, effects of different molecular diffusion models [9,10], initial gradient of fuel composition and strain rate [11,12,19, 20] on non-premixed ignition were also examined. For example, Knikker et al. [9] found that non-premixed ignition can be characterized by the propagation of reaction front in mixture fraction space. Mukhopadhyay and Abraham [14,15] demonstrated that this reaction front movement plays an important role in the ignition with initial composition stratification. Mastorakos [13] proposed that the propagation speed of reaction front is related to laminar flame speed of stoichiometric reactant mixture. However, the mechanism for the reaction front movement during non-premixed ignition is still not well understood. The connection between the initial ignition kernel and the following establishment of the whole flame has been rarely investigated. Furthermore, it is still not clear how the non-premixed

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Fig. 1. Schematic diagram of initial temperature and mixture fraction distributions. x_c denotes the center position of the diffusion layer and δ is the thickness of the diffusion layer.

ignition is affected by the initial thermal and composition gradient in the diffusion layer.

Hydrogen is widely adopted as a fuel booster due to its high reactivity and wide flammability range. Typical applications include the flameless and MILD combustion [1,21–24]. Dally et al. [21] experimentally investigated the hydrogen addition on the jet-in-hotco-flow. They found that hydrogen addition greatly enhances the auto-ignition within the diffusion layer between the fuel and oxidizer streams and that the stabilization mechanism changes even when a small amount of hydrogen is added [23–25]. Although different stabilization mechanisms for diffusion methane/hydrogen jet flames were observed in experiments, the mechanism of hydrogen enhancement on auto-ignition within the diffusion layer still remains unclear. Besides, the relative contributions of high reactivity and strong mobility of hydrogen to the non-premixed ignition have not been thoroughly studied.

Based on the above considerations, the objectives of this study are to assess the effects of hydrogen (H_2) addition on the ignition delay time of iso-octane (iC₈H₁₈) by hot air in a diffusion layer and to identify and interpret possible modes of reaction front propagation during non-premixed ignition. The emphasis is put on assessing and interpreting the influence of preferential mass diffusion of hydrogen on the non-premixed ignition process. Besides, the influence of diffusion layer thickness is also examined and the underlying mechanism is investigated.

2. Numerical model and specifications

We investigated the transient ignition process in a diffusion layer between cold fuel (H_2/iC_8H_{18} blends) and hot air. As shown in Fig. 1, all the mixture is initially static (i.e., $U_0 = 0 \text{ cm/s}$) with $P_0 = 1$ atm. On the left side of the diffusion layer is H_2/iC_8H_{18} blends at $T_{0,F} = 423$ K. The volume fraction of H_2 in H_2/iC_8H_{18} blends is denoted as α and it varies from 0 (pure i C_8H_{18}) to 1 (pure H_2). On the right side is hot air at $T_{0,0} = 1400$ K. The ignition delay time is defined as time when the maximum temperature reaches 1800 K. Besides, different ignition criteria [9] were used and the differences were found to be negligible. To quantify the local mixing level between fuel and oxidizer, we used the mixture fraction defined as [26,27]:

$$Z = \frac{Y_C/(\nu_C W_C) + Y_H/(\nu_H W_H) + 2(Y_{0,0} - Y_0)/(\nu_0 W_0)}{Y_{C,F}/(\nu_C W_C) + Y_{H,F}/(\nu_H W_H) + 2Y_{0,0}/(\nu_0 W_0)}$$
(1)

where Y_k denotes the mass fraction of *k*th element and υ_k is the coefficient in the global reaction $\upsilon_C C + \upsilon_H H + \upsilon_0 O \rightarrow$ Product. $Y_{O,O}$ is the mass fraction of oxygen element in air and Y_{CF} and $Y_{H,F}$ are

respectively the mass fraction of carbon and hydrogen elements on fuel side. The mass fraction of each element can be readily obtained in simulation and the stoichiometric mixture fraction can be used to indicate the reaction front movement in the mixture fraction space.

In the diffusion layer, the initial distributions of fuel and air were specified by the following expression for Z [9]:

$$Z = \frac{1}{2} \left[1 + erf\left(\frac{x - x_c}{\delta}\right) \right]$$
(2)

in which *erf* is an error function, x_c denotes the diffusion layer center, and δ is the diffusion layer thickness (see Fig. 1). The length of the computation field is 2 cm and x_c is at 0.6 cm. The mass fraction and enthalpy of each species, Y_k and h_k , were initialized according to $Y_k = Z \times Y_{k,F} + (1-Z) \times Y_{k,O}$ and $h_k = Z \times h_{k,F} + (1-Z) \times h_{k,O}$, respectively. The initial temperature can be calculated from enthalpy and concentration of all species [9].

It is noted that the above initial distribution assumes frozen flow without chemical reaction and equal diffusivity of all species during the formation of the diffusion layer. In fact, H₂ has much larger diffusivity than iC₈H₁₈. Therefore, we first considered the non-premixed ignition process with relatively small diffusion layer thickness so that the effect of differential mass diffusion between H₂ and iC₈H₁₈ can be examined. Since thicker diffusion layer helps to elucidate the modes of reaction front propagation and the underlying mechanism, we also considered ignition in a diffusion layer with a broad range of thickness, $\delta = 0-800 \,\mu\text{m}$. During the ignition process, oxygen (O₂) consumption rate ($\dot{\omega}_{02}$) was used as reactive region indicator. Furthermore, its maximum value, $\dot{\omega}_{02,max}$, was used to characterize the reaction front.

The transient non-premixed ignition process was simulated by the in-house code A-SURF. A-SURF solves the conservation equations for 1D, compressible, multi-component, reactive flow using the finite volume method. A-SURF has been successfully used in previous studies on ignition and flame propagation. The details on governing equations, numerical methods, and code validation for A-SURF can be found in Refs. [28,29] and thereby were not repeated here. In simulation, we used the detailed chemical mechanism developed for iso-octane by Chaos et al. [30]. This mechanism includes the sub-mechanism for hydrogen oxidation developed by Li et al. [31]. Therefore, it can be used to study the non-premixed ignition of H₂/iC₈H₁₈ blends. CHEMKIN packages [32] were incorporated into A-SURF to calculate the temperature- and componentdependent thermodynamic and transport properties as well as the reaction rates. The mixture-averaged model was used to evaluate the transport coefficients. The Soret diffusion were considered only for H₂ and H [37,38] since the Soret diffusion for heavier species is negligible. In simulation, dynamically adaptive mesh refinement was used and the diffusion layer was always covered by the finest meshes whose size is 2µm. The corresponding time step size is 0.8 ns. Numerical convergence was checked and ensured by further decreasing the time step and mesh size in simulation.

3. Results and discussion

We first considered the non-premixed ignition in the diffusion layer with nearly zero thickness (i.e., $\delta \approx 0$) and examined the effects of hydrogen addition on the non-premixed ignition delay time in Section 3.1. Then we studied the reaction front propagation and different ignition modes in Section 3.2. Finally, we assessed the influence of initial diffusion layer thickness on non-premixed ignition in Section 3.3.

3.1. Effects of hydrogen addition on the non-premixed ignition

Figure 2 shows the ignition delay time, $\tau_{ig,0D}$, for the 0D constant-pressure homogeneous ignition system whose initial



Fig. 2. Ignition delay time of adiabatic homogeneous system initialized by the conditions within the diffusion layer. The triangles denote the stoichiometric mixture fraction, Z_{st} . The circles denote the so-called most reactive mixture fraction $Z_{mr,0D}$. The squares denote $Z_{mr,1D}$ defined in the text.

conditions are extracted from the 1D diffusion layer and can be uniquely parameterized by Z. With the increase of Z, the composition of cold fuel increases in the mixture, and thereby the mixture temperature decreases while its equivalence ratio increases. Consequently, $\tau_{ig,0D}$ changes non-monotonically with Z. For each H₂ blending level, there is a minimum value of $\tau_{ig,0D}$ at the so-called most reactive mixture fraction $Z_{mr,0D}$ (circles in Fig. 2) [7,13] and the corresponding ignition delay time is denoted as $\tau_{ig,0D}'$. Figure 2 indicates that $Z_{mr,0D}$ decreases slightly and $\tau_{ig,0D}'$ decreases greatly with the increase of H₂ blending level. Mastorakos et al. [7] proposed that in 1D diffusion layer the initial ignition kernel (IIK) usually occurs at the position of most reactive mixture fraction $Z_{mr,0D}$. In 1D simulation, the initial ignition kernel occurs at the position whose mixture fraction is denoted as $Z_{mr,1D}$ (squares in Fig. 2). Due to mass and heat transport, $Z_{mr,1D}$ is slightly different from $Z_{mr,0D}$. Besides, unlike $Z_{mr,0D}$, $Z_{mr,1D}$ is shown to change significantly with H₂ blending level.

The 1D simulation results also indicate that reaction front first forms at $Z_{mr,1D}$ and then moves to $Z_{mr,0D}$ and stabilizes there. The difference between $Z_{mr,1D}$ and $Z_{mr,0D}$ has little influence on the ignition process. Therefore, only $Z_{mr,0D}$ is used in the following discussion. Besides, the movement of reaction front in the *Z* space reflects different ignition behaviors within the diffusion layer and it will be discussed later. Previous studies [7–9,14] showed that the non-premixed ignition can be generalized as the movement of reaction front from Z_{mr} to Z_{st} . However, the intrinsic behavior of the reaction front movement remains unclear, which will be interpreted in next sub-section.

Figure 3 shows the evolution of temperature and mixture fraction distributions in the physical space for $\alpha = 0.0$. It is seen that the mixing layer expands due to mass diffusion and heat conduction. The distance between $Z_{mr,0D}$ and Z_{st} increases with time in the physical space. The local temperature at $Z_{mr,0D}$ and Z_{st} decreases before ignition. Ignition occurs first near high temperature side due to the exponential dependence of reaction on temperature. Then the reaction region moves towards the fuel-rich side. Similar phenomenon was observed in previous studies [33].

Figure 4 shows the temporal evolution of scalar dissipation rate, χ in the mixture fraction space. It is shown that χ profile decays exponentially before ignition occurs. Heat released due to autoignition have strong influence on the χ -Z profile. For example,



Fig. 3. Distribution of mixture fraction (upper) and temperature (lower) in physical space at different ignition stages for pure iso-octane ($\alpha = 0.0$). x_c denotes the initial position of mixing layer.



Fig. 4. Temporal evolution of scalar dissipation rate in mixture fraction space at $\alpha = 0.0$ (upper) and $\alpha = 0.1$ (lower).

at $\alpha = 0.0$, it is seen that χ increases behind reaction region while it decreases ahead of the reaction region. This is due to the two competitive effects of heat release on χ -*Z*: gas expansion reduces χ while temperature increase promotes mass diffusivity. These results are consistent with the observation and discussion in [15]. However, at $\alpha = 0.1$, the monotonicity of χ -*Z* around reactive region disappears due to the high diffusivity of H₂.

Figure 5 plots the 1D non-premixed ignition delay time, $\tau_{ig, 1D}$, as a function of H₂ blending level (empty circles). It is observed that $\tau_{ig, 1D}$ first decreases exponentially with α for $\alpha < 0.4$ and then remains nearly constant for $0.4 < \alpha < 1.0$. This indicates that a small amount of H₂ blending can significantly promote the non-premixed ignition of iC₈H₁₈ by hot air. Such ignition promotion is mainly caused by the high mobility and reactivity of H₂. In Fig. 5, we also plot $\tau'_{ig,0D}$ for 0D homogenous ignition at $Z = Z_{mr,0D}$ (i.e., the circles in Fig. 2) and $\tau'_{ig,1D}$ for 1D non-premixed ignition with the mass diffusivity of H₂ being artificially modified to the value of iC₈H₁₈ (i.e., D_{H2} = D_{iC8H18}). Even for pure iso-octane (i.e., $\alpha = 0.0$), H₂ is produced around the ignition kernel and thereby changing the mass diffusivity of H₂ still affects the ignition delay for pure



Fig. 5. Change of ignition delay time with hydrogen blending level.

iso-octane with $\alpha = 0$ in 1D case. The large difference between $\tau'_{ig,1D}$ and $\tau_{ig,1D}$ demonstrates that the preferential mass diffusion of H₂ over iC₈H₁₈ significantly enhances the non-premixed ignition. This is mainly due to the fact that the high mobility of hydrogen molecule makes the local H₂ blending level at the initial ignition kernel (IIK) much higher than α specified at the boundary outside the diffusion layer. Similar phenomenon was also observed in the non-premixed ignition of H₂/CH₄ fuel blends [33].

When the effects of preferential mass diffusion of H₂ over iC_8H_{18} is eliminated by setting $D_{H2} = D_{iC8H18}$, the non-premixed ignition delay time $\tau'_{ig,1D}$ is shown in Fig. 5 to have similar trend as homogenous ignition delay time $\tau'_{ig,0D}$ at $Z_{mr,0D}$. The difference between $\tau'_{ig,1D}$ and $\tau'_{ig,0D}$ is due to the fact that the non-premixed ignition involves two processes: (1) nearly 0D homogeneous ignition at $Z_{mr,0D}$ and (2) the concomitant 1D transportation of fuel and heat into or out of the IIK. For the second process, mass and heat transport leads to opposite consequences: fuel transfer into the IIK promotes ignition while heat transfer out of the IIK inhibits ignition. Since the mass diffusivity of iC_8H_{18} is smaller than the heat diffusivity (i.e., Le > 1 for iC_8H_{18}), the inhibition due to heat loss dominates over the enhancement caused by fuel supply. Consequently, we have $\tau'_{ig,0D} < \tau'_{ig,1D}$ as shown in Fig. 5.

3.2. Modes of reaction front propagation during non-premixed ignition in a diffusion layer

To understand the details involved in the non-premixed ignition process, we tracked the reaction front propagation. The reaction front was defined as the location where local maximum O_2 consumption rate occurs. Double reaction fronts were identified since two local peaks appear in the distribution of O_2 consumption rate.

To justify the usage of $\dot{\omega}_{02,max}$ for reaction front identification, other indicators such as reaction rate of OH species, $\dot{\omega}_{OH}$ [11,20] and heat release rate, Q [11,12] are also tested. Figure 6 shows the reaction region indicated by these three quantities. It is shown that all these three quantities give consistent indication of reactive region position in *Z* space. For all the cases in the current work, only one reaction front is observed before ignition. We use the local maximum of $\dot{\omega}_{02,max}$ profile as reaction front indication because $\dot{\omega}_{02}$ is always negative and its local maximum clearly indicates the position of the reaction front.

Figure 7 shows the evolution of mixture fraction at the reaction front for different H_2 blending levels. The time was normalized



Fig. 6. Contour of O_2 reaction rate $\dot{\omega}_{02}$ (left), heat release rate Q (middle), OH reaction rate $\dot{\omega}_{0H}$ (right) at Z-t space for $\alpha = 0.1$.

by $\tau_{ig,1D}$ of each case so that the non-premixed ignition occurs at $t/\tau_{ig,1D} = 1$. The results for $D_{H2} = D_{iC8H18}$ (i.e., the mass diffusivity of H₂ were artificially modified) and those for $D_{H2} \neq D_{iC8H18}$ (i.e., without modifying the mass diffusivity of H₂) were plotted together for comparison. For pure iC_8H_{18} (i.e., $\alpha = 0$) as shown in Fig. 7a, the reaction front first initiates at $Z_{mr,1D}$ and then immediately moves to $Z_{mr.0D}$. It stabilizes there until ignition occurs. Once ignition occurs, the reaction front splits into two branches: a stoichiometric branch around Z_{st} and a fuel-rich branch. The fuel-rich branch forms due to the leakage of small amount of O₂ into fuel side after its inadequate reaction at stoichiometry. Similar observation was also reported in [9]. The stoichiometric branch stands for the typical diffusion flame and the fuel-rich branch represents a rich premixed flame. The fuel-rich branch extinguishes eventually while the stoichiometric branch lasts for a longer duration. It is noted that for $\alpha = 0$, the reaction front mainly stays around $Z = Z_{mr,0D}$ before $t = 0.9\tau_{ig,1D}$ (point A in Fig. 7a). There is little fuel transported into reaction front and thereby the local mixture fraction remains nearly constant. This indicates that the ignition process is mainly dominated by the local autoignition at $Z_{\rm mr,0D}$. Such reaction front propagation is referred to as the reaction-driven mode. In this mode, reaction front moves slowly or even statically in Z space. Its movement is mainly driven by local or sequential homogeneous autoignition.

However, for $\alpha = 0.1$ and $\alpha = 0.4$ with $D_{H2} \neq D_{iC8H18}$, reaction front propagates in a different behavior. For $\alpha = 0.1$, the reaction front starts to move away from $Z = Z_{mr,0D}$ around $t = 0.35\tau_{ig,1D}$ (point D in Fig. 7b). During $0.35\tau_{ig,1D} < t < \tau_{ig,1D}$, due to fuel diffusion the fuel concentration at the reaction front increases and so does Z. After ignition, only one stoichiometric branch of reaction front is observed since H₂ consumes O₂ which is supposed to penetrate into the fuel side. Similarly, for $\alpha = 0.4$, the reaction front starts to move away from $Z = Z_{mr,0D}$ around $t = 0.5\tau_{ig,1D}$ and finally reaches Z_{st} . Therefore, with hydrogen addition the reaction front movement is in the diffusion-driven mode, in which mass diffusion and heat transfer play an important role. In diffusion-driven mode, the local mixture fraction at the reaction front starts to increase at an early stage and its movement in Z space is mainly driven by fuel diffusion and heat transfer.

For $\alpha = 0.1$ and $\alpha = 0.4$ with $D_{H2} = D_{iC8H18}$ (open circles in Fig. 7b and c), the reaction front is shown to mainly stay around $Z = Z_{mr,0D}$ before $t = 0.9\tau_{ig,1D}$. This is similar to the case without hydrogen addition (solid circles in Fig. 7a). Given the fact that the normalization factor, $\tau_{ig,1D}$ greatly decreases with α , hydrogen addition triggers the transition of reaction front movement from reaction-driven mode to diffusion-driven mode at a very early



Fig. 7. Evolution of mixture fraction at the reaction front. The open circles are for cases in which the mass diffusivity of H_2 being artificially modified to the value of iC_8H_{18} (i.e., $D_{H2} = D_{iC8H18}$); and the closed circles are for cases without modifying the mass diffusivity of H_2 (i.e., $D_{H2} \neq D_{iC8H18}$). The solid lines stand for Da = 1 (above the solid line: Da < 1); below the solid line: Da > 1). The horizontal dashed and dash-dotted lines denote Z_{st} and $Z_{mr,0D}$, respectively.

stage. Therefore, the preferential mass diffusion of H_2 over iC_8H_{18} plays an important role in the mode of reaction front propagation.

The reaction front propagation mode can also be determined by the Damköhler number, $Da = \tau_{diff} / \tau_{chem}$, which is the ratio between the characteristic time for diffusion, au_{diff} , and that for chemical reaction, τ_{chem} . The basic idea is: ignition is equivalent process of reaction front propagation from Z_{mr} to Z_{st} , and τ_{diff} and τ_{chem} denotes the time that needed to drive reaction front to reach Z_{st} by diffusion effect or reaction, respectively. The diffusion time was defined as $1/\chi$ where χ is the local scalar dissipation rate. The characteristic reaction time was defined as $\tau_{chem} = (D/\chi)^{1/2} / S_{d,0}$. *D* is the mass-averaged diffusion coefficient. The numerator $(D/\chi)^{1/2}$ represents an estimate of the physical distance between local Z and Z_{st} , and the denominator $S_{d,0}$ represents reaction front movement speed solely driven by sequential autoignition. This speed can be pre-computed by homogeneous reaction system as $S_{d,0} = dx/d\tau_{ig,0D} = (dx/dZ)/(d\tau_{ig,0D}/dZ)$. The expression of *Da* reduces to $Da = (dZ/d\tau_{ig,0D})/\chi$. Both τ_{diff} and τ_{chem} change with position and time, thus Damköhler number varies in space and time.

The time evolution of Damköhler number in mixture fraction space is shown in Fig. 8 for pure iso-octane ($\alpha = 0.0$). For small *Z*, Da > 1 (i.e., $\tau_{che} m < \tau_{diff}$) and the reaction front propagation in this region is driven by local or sequential autoignition. On the contrary side, Da < 1 for large *Z* and thereby the reaction front propagation in this region is driven by fuel diffusion and heat transfer.

The solid lines in Fig. 7 denote *Z* position of Da = 1 as function of time: above this line is the diffusion-driven mode with Da < 1;



Fig. 8. Time evolution of Damköhler number in mixture fraction space of $\alpha = 0.1$ at different ignition stages.

and below this line is the reaction-driven mode with Da > 1. For pure iso-octane ($\alpha = 0$), the reaction front trajectory falls in Da > 1region for most of time before ignition. This indicates that the ignition is mainly dominated by autoignition. At around $t = 0.9\tau_{ig,ID}$, the reaction front suddenly moves towards Z_{st} , and switches into



Fig. 9. Distributions of heat release rate (Q, pink dashed lines) and two parts of displacement speed ($S_{d,chem}$, solid red lines; and $S_{d,diff}$, solid blue lines) within the diffusion layer. Each row corresponds to a single value of α . These subfigures correspond to the points, A-I, marked in Fig. 7. Chemical displacement speed $S_{d,chem}$ is always negative due to the consumption of O₂. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the diffusion-driven mode with Da < 1. For the case with hydrogen addition ($\alpha = 0.1$), the region occupied by reaction-driven mode shrinks and the reaction front switches into the diffusion-driven mode earlier than the case of $\alpha = 0$. When the H₂ blending ratio is further increase to $\alpha = 0.4$, the region of reaction-driven mode become much narrower. Diffusion-driven mode becomes more dominated during the ignition process.

To further characterize the ignition mode, we used the displacement speed, S_d , based on oxygen [34,35]:

$$\mathbf{S}_{\mathbf{d}} = \frac{1}{\boldsymbol{\rho}_{\mathbf{u}} |\nabla Y_{02}|} \left[\underbrace{\underbrace{\boldsymbol{\rho}} \dot{\boldsymbol{\omega}}'_{\mathbf{02}}}_{reaction} + \underbrace{\frac{\partial}{\partial x} \left(\boldsymbol{\rho} \boldsymbol{D}_{\mathbf{02}} \frac{\partial \mathbf{Y}_{\mathbf{02}}}{\partial x} \right)}_{diffusion} \right]$$
(3)

Eq. (3) indicates that S_d consists of two parts: one corresponds to chemical reaction, S_{chem} ; and the other corresponds to diffusion, S_{diff} (i.e., $S_d = S_{chem} + S_{diff}$). Although S_d analysis has been

used mostly for premixed flames, it's still applicable in the premixed region within the nonpremixed mixing layer. The reaction front propagation mode can also be characterized by the ratio between S_{chem} and S_{diff} : it is dominated by chemical reaction when $S_{chem} > S_{diff}$; and it is dominated by diffusion process when S_{chem} is in the same order of magnitude as S_{diff} .

Figure 9 shows the distribution of S_{chem} , S_{diff} and heat release rate Q for points A-I marked in Fig. 7. S_{chem} is always negative since O_2 is consumed and it makes negative contribution to the total displacement speed. Positive S_{diff} means that O_2 diffuses into fuel side and vice versa. For point A, S_{chem} is one order of magnitude larger than S_{diff} . This indicates that chemical reaction has dominant contribution to the total displacement speed, and thereby the reaction front is in the reaction-driven mode. Both S_{chem} and S_{diff} are relatively small due to the low reactivity of pure iso-octane. When the reaction front switches into the diffusion-driven region at point B, both S_{chem} and S_{diff} increase exponentially and they have the same order of magnitude, indicating the balance between diffusion and reaction. Therefore, the reaction front can be considered as a premixed flamelet moving in the diffusion layer of fuel and oxidizer. It is observed that the heat release rate at point B is one order of magnitude larger than that at point A. For cases with small amount of H₂ addition, at point D S_{chem} is much larger than S_{diff} , indicating that chemical reaction dominates over diffusion. After reaction front goes into diffusion-driven mode at point E, S_{chem} and S_{diff} are in the same order of magnitude. Similar results are obtained for $\alpha = 0.4$ at points G and H. For the cases with D_{H2} = D_{iC8H18} (points C, F, and I), both Q and S_{diff} remain nearly unchanged while S_{chem} increases.

For generality, we also test the *Da* criteria at higher ambient pressure of 20 and 40 atm with other parameters unchanged (fuel temperature $T_{F,0}$ is 50 K over the corresponding boiling point). The results at high pressures are consistent with those at $P_0 = 1$ atm. Both *Da* criteria and *S_d* analysis gives consistent identification of the reaction front mode.

It should be noted that in the definition of *Da*, we use a series of homogeneous adiabatic reactor to calculate $d\tau_{ig,0D}/dZ$. However, the preferential diffusion of H₂ into the hot air side prior to ignition causes $d\tau_{ig,0D}/dZ$ to deviate from its original value. Considering good consistency between *Da* criteria with *S*_d analysis, approximating $d\tau_{ig,0D}/dZ$ by a series of homogeneous adiabatic reactor seems to be reasonable.

In brief summary, for the non-premixed ignition in a diffusion layer, the reaction front propagation has two modes: one is the reaction-driven mode characterized by local or sequential homogeneous autoignition, and the other is the diffusion-driven mode which depends on the balance of chemical reaction and diffusion. The analysis based on Damköhler number and consumption speeds yields consistent results on the mode of reaction front propagation. The reaction-driven mode predominates at the beginning and the diffusion-driven mode appears when ignition happens (i.e., $t \approx \tau_{ig,1D}$).

3.3. Influence of diffusion layer thickness on the non-premixed ignition

Previous studies [7,11,12,14,36] reported that the ignition delay time depends on the initial gradient of composition and temperature. In this sub-section, we examined the influence of diffusion layer thickness on non-premixed ignition delay time and different ignition modes.

To investigate the dependence of $\tau_{ig,1D}$ on initial mixing layer thickess, we use initial characteristic Damköhler number, Da_0 , to describe the normalized initial mixing layer thickness. Da_0 is defined as $Da_0 = 1/(\chi_{st,0} \overline{d\tau_{ig,0D}/dZ})$, where $\chi_{st,0}$ is the initial scalar dissipation rate at Z_{st} ; $\overline{d\tau_{ig,0D}/dZ}$ is calculated as $(\tau_{st,0D}-\tau'_{ig,0D})/(Z_{st}-Z_{mr})$, where $\tau_{st,0D}$ and $\tau'_{ig,0D}$ are the homogeneous ignition delay time at Z_{st} and $Z_{mr,0D}$ (as shown in Fig. 2), respectively. The definition of Da_0 is consistent with Da that we used in Section 3.2, except that Da_0 can be obtained before simulation. For each H₂ blending level, $d\tau_{ig,0D}/dZ$ is fixed, increasing δ will decrease $\chi_{st,0}$ and thus increase Da_0 . Therefore, Da_0 is used to indicate the normalized mixing layer thickness.

Figure 10 shows the change of the normalized non-premixed ignition delay time, $\tau_{ig,1D} / \tau'_{ig,0D}$ and $\tau'_{ig,1D} / \tau'_{ig,0D}$, with initial characteristic Damköhler number, Da_0 at different H₂ blending levels of $\alpha = 0$, 0.1, 0.4 and 0.8. It is shown that, when the mass diffusivity of H₂ is not artificially modified (i.e., $D_{H2} \neq D_{iC8H18}$), $\tau_{ig,1D}$ decreases with Da_0 for pure iC₈H₁₈ (i.e., $\alpha = 0$); while it increases with Da_0 for H₂/iC₈H₁₈ blends (i.e., $\alpha > 0$). This indicates that H₂ addition can change the underlying mechanism for non-premixed ignition of iC₈H₁₈ in a diffusion layer. It is also shown that $\tau_{ig,1D} / \tau'_{ig,0D}$ approximates to unity when Da_0 increases



Fig. 10. Nonpremixed ignition delay time normalized by minimum adiabatic homogeneous ignition delay time, $\tau'_{ig,0D}$ of each H₂ blending level, plotted against initial Damköhler number, $Da_0=1/(\chi_{st,0} d\overline{\tau}_{ig,0D}/d\overline{Z})$. Diffusivity of H₂ is (a) unchanged and (b) artificially damped as $D_{H2} = D_{iC8H18}$. Dashed lines denote different mixing layer thickness. Vertical line denotes $Da_0 = 0.1$.

since the ignition becomes reaction-driven mode. When the mass diffusivity of H₂ is artificially modified to the value of iC₈H₁₈ (i.e., $D_{H2} = D_{iC8H18}$), Fig. 10b shows that $\tau'_{ig,1D}/\tau'_{ig,0D}$ always decreases with δ and that small amount of H₂ blending (i.e., $\alpha < 0.4$) has little effects on $\tau'_{ig,1D}$. Therefore, these results further demonstrate that the preferential mass diffusion of H₂ over iC₈H₁₈ greatly affects the non-premixed ignition in a diffusion layer.

The different trends shown in Fig. 10 are mainly due to the dependence of ignition on the fuel mass diffusivity and heat transfer. For pure iC_8H_{18} or the cases of $D_{H2} = D_{iC8H18}$, the fuel has relatively low mass diffusivity and the non-premixed ignition is controlled by local chemical reaction around $Z = Z_{mr,0D}$ (as shown by Fig. 7a). Consequently, larger gradient (i.e., smaller Da_0 and δ) results in greater heat loss from the ignition kernel and thereby the non-premixed ignition delay time decreases with Da_0 . For cases with H_2 addition, the mass diffusivity of H_2 is much higher than heat conduction. Before ignition, the reaction front does not always remain around $Z = Z_{mr,0D}$ and it is greatly affected by diffusion (as shown by Fig. 7b and c). Since larger gradient (i.e., smaller Da_0 and δ) results in more fuel transported into the ignition kernel, $\tau_{ig. 1D}$ increases with δ for H_2/iC_8H_{18} blends (i.e., $\alpha > 0$).

It is noted that H_2 addition has the most significant effect on non-premixed ignition within the region of $Da_0 < 0.1$. This is because H_2 addition becomes significant at small δ when diffusion effect is strong enough to couple with the chemistry reaction, leading to diffusion-driven ignition. Since we use a planar mixing layer, ignition always occurs and infinitesimal Da doesn't imply extinction. For large Da_0 , chemistry reaction dominates over diffusion effect, and the chemical effect of H_2 on ignition is limited due to its small molecular weight. Therefore, the criteria of $Da_0 < 0.1$ can be used to delineate the effect of H_2 addition in terms of its diffusion effect.

To quantitatively demonstrate the above explanation, in Fig. 11 we plotted the evolution of local temperature and mixture fraction at the reaction front with different initial mixing layer thickness, δ . The left and right column in Fig. 11 represents for $\alpha = 0$ and $\alpha = 0.1$, respectively. For $\alpha = 0$, we also plotted the local temperature at $Z=Z_{mr,0D}$ (see the dash lines). For pure iC₈H₁₈ (i.e.,



Fig. 11. Evolution of temperature and mixture fraction at the reaction front for $\alpha = 0$ (left) and $\alpha = 0.1$ (right). The temperature for 0D homogenous ignition at $Z_{mr,0D}$ is also presented as dash-dotted lines. Besides, for $\alpha = 0$ the local temperature at $Z = Z_{mr,0D}$ of 1D simulation is shown as dashed lines.

 $\alpha = 0$), with the increase of δ , a discontinuity in the temperature trajectory at reaction front appears. However, the evolution of T at $Z = Z_{mr,0D}$ is continuous, indicating that the local autoignition at $Z = Z_{mr,0D}$ rather than that at reaction front, dominates the nonpremixed ignition. Therefore, when the diffusion layer thickness is small, strong heat loss leads to the lower initial temperature at $Z = Z_{mr,0D}$ leads to slower ignition. With the increase of diffusion layer thickness, the local temperature at $Z = Z_{mr,0D}$ becomes higher due to less heat loss and thereby the ignition delay time $\tau_{ig, 1D}$ decreases with δ for $\alpha = 0$. However, for iC₈H₁₈ with H₂ addition (i.e., $\alpha = 0.1$), the evolution of T and Z at reaction front are always continuous at different values of δ . For this case the non-premixed ignition is controlled by fuel diffusion. When the diffusion layer is small, large composition gradient transport relatively more H₂ into ignition kernel and promotes ignition. The larger the diffusion layer thickness, the smaller the fuel concentration gradient and the less the amount of fuel diffuses into the ignition kernel. Consequently, the ignition delay time $\tau_{ig, 1D}$ increases with δ for $\alpha = 0.1$. Figure 11 also shows the evolution of temperature for 0D homogeneous ignition with initial condition specified at $Z = Z_{mr,0D}$ (dashdotted lines). With the increase of diffusion layer thickness, the transport due to gradient becomes weaker and thereby 1D nonpremixed ignition approaches to 0D homogeneous ignition.

4. Conclusions

Numerical simulations were conducted to investigate the transient non-premixed ignition within a diffusion layer between iC_8H_{18}/H_2 and hot air. Detailed chemistry and transport were considered in simulation. It was found that small amount of H_2 addition greatly reduced the ignition delay time. This is mainly caused by the high preferential mass diffusivity of H_2 . Due to the preferential mass diffusion of H_2 over iC_8H_{18} , the local H_2 blending level at the initial ignition kernel is in fact much higher than that at the boundary. The ignition within the diffusion layer consists of two processes: (1) homogeneous ignition at the initial ignition kernel which is located at the position of most reactive mixture fraction; and (2) diffusion transportation of fuel into and heat out of the ignition kernel.

Two modes of reaction front propagation were identified through the analysis based on Damköhler number and consumption speeds: one is the reaction-driven mode characterized by local or sequential homogeneous autoignition and the other is the diffusion-driven mode, which depends on the balance of mass diffusion, heat transfer and chemical reaction. Hydrogen addition makes the reaction front switch from reaction-driven mode to diffusion-driven mode.

Furthermore, it was found that the diffusion layer thickness greatly affects the non-premixed ignition delay time. For pure iC_8H_{18} , the ignition delay time decrease with the thickness; while the opposite trend happens to H_2/iC_8H_{18} blends. This is due to the fact that for pure iC_8H_{18} with low mass diffusivity, the non-premixed ignition process is dominated by the local homogeneous autoignition at the most reactive mixture fraction. When hydrogen with high mass diffusivity is added, the non-premixed ignition is controlled by fuel diffusion: the larger the diffusion layer thickness, the less the amount of fuel diffused into the ignition kernel and thereby the longer the ignition delay time.

It is noted that in the present study, normal pressure and a high temperature of oxidizer of 1400 K are considered. Further study is needed for conditions at higher pressures and lower oxidizer temperatures. At such conditions, the low-temperature chemistry plays an important role in the ignition process. There might be competition/interaction between low-temperature ignition and high-temperature ignition, especially for fuels like n-heptane with strong negative temperature coefficient behavior.

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