Prediction of ignition modes of NTC-fuel/air mixtures with temperature and concentration fluctuations

Minh Bau Luong^{a,*}, Francisco E. Hernández Pérez^a, Hong G. Im^a

^a*Clean Combustion Research Center, King Abdullah University of Science and Technology, Thuwal, Saudi Arabia*

Abstract

The ignition regime criteria proposed by Im et al. [1] are extended to account for temperature and concentration fluctuations. The newly-developed criteria are applied to negative temperature coefficient (NTC) and non-NTC fuels. The statistical volume-averaged Sankaran number, $\bar{Sa}$, and the volumetric fraction of $Sa < 1$, $F_{Sa,S}$, are proposed as new metrics to determine *a priori* the combustion modes, combustion intensity, and ignition delay times, with $F_{Sa,S}$ being used to quantify the heat release fraction from spontaneous ignition. It is demonstrated that $F_{Sa,S}$ agrees acceptably with the fraction of heat release from spontaneous ignition obtained by the Damköhler-based analysis from direct numerical simulation (DNS) data. The ignition modes can also be predicted by $\bar{Sa}$ regardless of the NTC and non-NTC characteristics of fuels over a wide range of initial mean temperatures and different fluctuation levels. Besides, the magnitude of $\bar{Sa}$ can be used to estimate $F_{Sa,S}$ due to its strong correlation with $F_{Sa,S}$. Additionally, the predicted Sa by the theory, Sa_p , is compared with the statistical mean $\bar{Sa}$ showing a consistent agreement and they are found to correlate with the combustion intensity that is characterized by the maximum heat release rate. Finally, the ignition delay time can be correlated with Sa_p for single-stage fuels, and for NTC fuels if the initial mean temperature lies outside the NTC regime.

Keywords: Direct numerical simulation (DNS), Ignition regimes, Temperature fluctuations, Concentration fluctuations, Negative temperature coefficient (NTC)

1. Introduction

Advanced low-temperature-combustion (LTC) strategies of downsized and boosted engines are capable of offering ultra-low emissions and higher efficiencies [2–4]. In these engines, combustion is chemically driven by the autoignition process with no direct means to control the ignition timing and combustion rate. The autoignition process and its timing are highly sensitive to the fuel types and operating conditions, including the intake temperature and pressure, thermal and compositional inhomogeneities of the mixture, the amount of exhaust gas recirculation (EGR), and cooling. As such, these engines are prone to suffer from pre-ignition, a higher possibility of knock, and even super-knock, which is characterized by high-pressure peaks and

oscillations leading to a severe structural damage [5–7]. Therefore, a reliable prediction of such abnormal ignition phenomena is of critical importance.

To reduce an excessive heat release rate (HRR), some levels of thermal and compositional fluctuations are desired to achieve a smooth combustion process [2, 3, 8, 9]. In the presence of thermal and/or concentration fluctuations, T' and ϕ' , a mixed combustion mode of spontaneous ignition and deflagration is encountered, leading to smooth and prolonged combustion with a lower peak pressure rise rate (PRR) [10–21].

Previous studies reported that larger T' leads to a predominant deflagration mode with a longer duration of HRR [22–26]. However, this effect is significantly reduced when the initial mean temperature, T_0 , falls into the negative temperature coefficient (NTC) regime due to the reduced ignition delay sensitivity to temperature [27–31]. Zhang et al. [32] used dimethyl ether (DME) as a surrogate

*Corresponding author.

Email address: minhbau.luong@kaust.edu.sa (Minh Bau Luong)

Nomenclature

CHRR	Cumulative heat release rate
DNS	Direct numerical simulation
HCCI	Homogeneous-charge compression ignition
HTR	High-temperature regime
ITR	Intermediate-temperature regime
LTR	Low-temperature regime
NTC	Negative temperature coefficient
NC (UC)	Negatively-correlated (uncorrelated)
RMS	Root mean square
SI (WI)	Strong (weak) ignition
SCCI/RCCI	Stratified-charge/reactivity-controlled compression ignition
PDF	Probability density function
PRF	Primary reference fuel

Symbols

Da	Damköhler number
$F_{\text{Sa},S}$	Volumetric fraction of $\text{Sa} < 1$
$F_{\text{Da},S}$	HRR fraction by the spontaneous ignition
l_e, l_T, l_ϕ	Integral length scale of turbulence, temperature, equivalence ratio
T' & ϕ'	RMS of T and ϕ
Sa	Sankaran number
Sa_p	Predicted Sankaran number
$\bar{\text{Sa}}$	Statistical mean Sa
$\bar{\text{Sa}}_n$	Normalized statistical mean $\bar{\text{Sa}}$, $\bar{\text{Sa}}_n = \bar{\text{Sa}}/(\bar{\text{Sa}}+1)$
$\text{Sa}_{p,n}$	Normalized predicted Sa_p , $\text{Sa}_{p,n} = \text{Sa}_p/(\text{Sa}_p+1)$
S_L	Laminar flame speed
S_{sp}	Spontaneous front speed
$T, p, \text{ and } \phi$	Temperature, pressure, and equivalence ratio
$T_0, p_0, \text{ and } \phi_0$	Initial mean $T, p, \text{ and } \phi$
τ_{ig}	Ignition delay time
$\tau_t (\tau_f)$	Turbulent (flame) time scale
u'	Turbulent velocity fluctuation

for NTC fuels to study the influence of the wavelength of the temperature fluctuation representing the integral length scale, the level of temperature gradients, and T_0 on the amplitude of pressure oscillation. They found that a larger wavelength of the temperature fluctuation and/or T_0 in the NTC regime induce a greater amplitude of the PRR oscillation. Pan et al. [33] also reported that the ignition-front of the cases inside the NTC regime travels faster than that of the cases outside the NTC regime, leading to increased interactions between the pressure wave and the developing reaction fronts, which in turn facilitates the formation of a detonation wave.

In a direct numerical simulation (DNS) study, Sankaran et al. [22] showed that the autoignition of a H_2 /air mixture with different T' was accurately predicted by a nondimensional Sankaran number, Sa , which is proportional to the ratio of laminar flame speed to the thermal-gradient characterized spontaneous propagation speed. In both experimental and computational studies, Sa was demonstrated as an accurate predictive criterion of the strong/weak mode for homogeneous mixtures in the presence of thermal nonuniformities [34–38]. Strozzi et al. [34] reported a satisfactory agreement between a quantitative Sa -based prediction of the occurrence of autoignition fronts and deflagrations and the chemiluminescence images in a rapid compression machine (RCM) during the early and intermediate stages of combustion. Mansfield et al. [35, 36] conducted experimental studies to investigate the ignition behavior of syngas and *iso*-octane in a RCM, reporting that Sa accurately predicts the location of the strong ignition limit for various equivalence ratios and locations.

Based on this theoretical framework, Im et al. [1] proposed a scaling analysis to account for the turbulent effect on the ignition process, and derived the turbulent ignition regime diagram to identify whether a combustion process is ignition controlled or flame-propagation controlled. The ignition regimes were classified into three categories: (1) weak ignition with the dominant mode of deflagration, (2) reaction-dominant strong ignition, and (3) mixing-dominant strong ignition. A further DNS study was conducted to verify the ignition regime diagram using syngas [39], obtaining consistent results with the predictions by the ignition regime diagram. In the previous studies [1, 39], the ignition criterion was qualitatively validated by using single-stage-ignition fuels with thermal fluc-

tuations only. Validation for NTC fuels with temperature and concentration fluctuations along with more quantitative assessment are needed.

In other contexts, Grogan et al. [40, 41] also proposed an ignition regime diagram specifically relevant to the shock tube and rapid compression facilities, while Bradley and co-workers [42–44] identified criteria to develop detonation from a hot spot, which have been further extended to other fuels by Chen and co-workers [45–47], yet the crucial effect of thermal stratification on autoignition and detonation development was recently underscored by Sow et al. [48].

While fundamental studies abound for different fuels with temperature and composition fluctuations, there has been little attempt at formulating a unified theory to predict the autoignition behavior in terms of a few key nondimensional numbers characterizing the nature of the bulk mixture inhomogeneities, especially with complex fuels exhibiting low temperature ignition chemistry, manifested as the NTC behavior, and simultaneous temperature/composition fluctuations. To this end, the objective of the present study is twofold: (1) to extend the ignition regime theory and diagram proposed by Im et al. [1] to accommodate both temperature and concentration fluctuations, and validate the theory for mixtures with NTC fuels, and (2) to derive a quantitative metric for the tendency of strong ignition in terms of the global quantities that characterize the mixture inhomogeneities. The main emphasis is on comparing and assessing various statistical quantities in the prediction metric, for mixtures with different fuels and conditions. New and existing DNS data are used in the analysis and discussion. In what follows, the theoretical framework is presented, and the results are described and discussed.

2. Theoretical framework

2.1. Ignition regime criteria

In this section, the turbulent ignition regime theory [1] is briefly summarized and further extended to conditions with both temperature and concentration fluctuations. According to Zeldovich’s theory [49], the spontaneous propagation speed of an ignition front, S_{sp} , is expressed as

$$S_{sp} = |\nabla \tau_{ig}|^{-1}, \quad (1)$$

where τ_{ig} is the homogeneous ignition delay time at a local mixture condition. While the original Zeldovich theory was derived in terms of temperature variations in the mixture only, it can be extended by expressing $\tau_{ig}(T, \phi)$ as a function of local temperature and equivalence ratio. In the presence of large spatial variations in τ_{ig} , local ignition establishes a deflagration front (weak ignition). For a mixture with a smaller level of τ_{ig} variations, the mixture ignites sequentially and S_{sp} exhibits a rapid ignition front propagation (strong ignition). If S_{sp} is sufficiently large and comparable to the speed of sound, the combustion wave may be coupled with the acoustic wave, leading to a rapid pressure rise and detonation development in some extreme conditions. In the asymptotic limit of zero gradient, S_{sp} becomes infinity, leading to homogeneous autoignition and involving an extremely high-pressure rise rate.

Based on DNS data, Sankaran et al. [22] proposed a criterion to determine the weak or strong ignition modes, introducing the Sankaran number (Sa), which is defined as

$$Sa = \beta \frac{S_L}{S_{sp}}, \quad (2)$$

where β is a weighting factor of 0.5 to account for the quantitative behavior observed in the DNS data. $Sa = 1$ serves as the boundary between the strong ($Sa < 1$) and the weak ($Sa > 1$) ignition.

In practical combustion systems, detailed spatially resolved information about the scalar fluctuations is not available, and thus, prediction of the ignition characteristics must be made based on the statistical averages or filtered quantities. Im et al. [1] conducted a theoretical scaling analysis to derive a refined criterion to predict the ignition regimes *a priori* for a mixture with turbulent velocity and temperature fluctuations. With a uniform composition, the gradient of ignition delay time, $\nabla\tau_{ig}$, is solely due to temperature variations and can be re-expressed using the chain rule as

$$|\nabla\tau_{ig}| = \left| \frac{d\tau_{ig}}{dT} \nabla T \right|, \quad (3)$$

where $d\tau_{ig}/dT$ represents the ignition delay sensitivity to temperature, and ∇T is the temperature gradient of the bulk mixture [1, 39]. In terms of the statistical average in turbulent conditions, Sa is approximated as

$$Sa \approx \beta S_L \left| \frac{d\tau_{ig}}{dT} \right| |\widetilde{\nabla T}|, \quad (4)$$

where S_L and τ_{ig} are evaluated at the initial bulk mixture conditions, and $|\widetilde{\nabla T}| \sim T'/\lambda_T$ denotes the statistical mean temperature gradient, estimated based on the variance of the temperature fluctuations and the Taylor mixing scale. Further scaling analysis yields the expression for the *predicted* Sankaran number, Sa_p , [1] as follows:

$$Sa_p = K Da_l^{-1/2}, \quad K = \beta \frac{1}{(\tau_f \tau_{ig})^{1/2}} \left| \frac{d\tau_{ig}}{dT} T' \right|, \quad (5)$$

where $Da_l = \tau_t/\tau_{ig}$ is Damköhler number, defined as the ratio of the turbulence integral time scale, l/u' , to the ignition delay time, and $\tau_f = \alpha/S_L^2$ is the characteristic flame time, where α is the thermal diffusivity of the initial bulk mixture.

Note that Sa_p is determined based on the statistical quantities of the mixture mean conditions, to be distinguished from the *exact* Sankaran number, Sa (Eq. 2), which is determined locally from the fully resolved information.

The turbulent ignition regime analysis is now further extended to account for both temperature and concentration fluctuations. In the presence of both temperature and equivalence ratio fluctuations, τ_{ig} is a function of T and ϕ . Using the chain rule, it follows that

$$|\nabla\tau_{ig}| = \left| \frac{\partial\tau_{ig}}{\partial T} \nabla T + \frac{\partial\tau_{ig}}{\partial\phi} \nabla\phi \right|, \quad (6)$$

where ∇T and $\nabla\phi$ are the temperature and equivalence ratio gradients of the local mixture [1]. Assuming that the temperature and composition fields are related to the turbulent flow field and, thus, have a comparable integral length scale, $l_T \sim l_\phi \sim l_e$, and following a similar approximation for $\widetilde{\nabla\phi} \sim \phi'/\lambda_T$ as in the temperature case, Eq. 5 is extended by determining the K factor as:

$$K = \beta \frac{1}{(\tau_f \tau_{ig})^{1/2}} \left(\left| \frac{\partial\tau_{ig}}{\partial T} T' \right| + s \left| \frac{\partial\tau_{ig}}{\partial\phi} \phi' \right| \right), \quad (7)$$

where s is the sign which becomes positive or negative depending on the statistical distribution of T and ϕ fluctuations. When s is positive if T and ϕ fluctuations are uncorrelated, i.e. T' and ϕ' are completely random, both quantities would enhance the sensitivity of τ_{ig} , thus promoting the deflagrative regime of ignition. On the other hand, if T' and ϕ' are negatively correlated, on statistical average one offsets the other, such that the two effects on K

become subtractive, hence s adopts a negative (positive) sign if $\partial\tau_{ig}/\partial T\partial\tau_{ig}/\partial\phi$ is positive (negative). Note that in PPC engines, a late direct injection induces a negatively correlated T - ϕ distribution and a nearly identical integral length scale, $l_T \sim l_\phi$, due to the cooling effect of fuel vaporization as verified by experimental studies using optical measurements [50–55], and by computational studies using DNS [56] and LES data from a full-cycle IC-engine simulation [57]. As such, the selection of T_0 , p_0 , and ϕ_0 , and the RMS values, T' and ϕ' , listed in Table 1 follows the previous studies [50–52, 56, 57].

2.2. Quantitative analysis

All reactant mixtures with temperature and composition fluctuations exhibit both weak and strong ignition behavior. If the DNS data are available, the exact distribution of Sa within the domain can be computed and the resulting ignition characteristics in terms of fractional contributions of weak and strong ignition to the total heat release can be determined. Such information serves as the reference target quantity. Here, the objective is to assess whether the Sa_p criterion based on the statistical mean can predict the ignition behavior consistently, for different fuels and temperature/composition fluctuations.

Figure 1 shows a representative DNS data field illustrating the instantaneous spatial distribution of (a) temperature and (b) equivalence ratio. At such initial conditions, the distribution of the corresponding local τ_{ig} and Sa are shown in (c) and (d), respectively. Subsequently, the probability density function (PDF) for Sa , $P(Sa)$ is constructed in Fig. 2, from which the fractional volume that would lead to strong (SI) and weak (WI) ignition behavior can be predicted. The volume fraction of the regions with $Sa < 1$, $F_{Sa,S}$, is given by

$$F_{Sa,S} = \int_0^1 P(Sa) dSa, \quad (8)$$

which is proposed as a predictive indicator of the fractional heat release rate associated with strong ignition. It follows that the contribution of the weak ignition (deflagration) is determined as $F_{Sa,W} = 1.0 - F_{Sa,S}$. If $F_{Sa,S}$ approaches unity, strong ignition is dominant. These quantities will be used to assess the accuracy of the predictive Sankaran number, Sa_p , in the next section. Note that the local mixtures with a very large τ_{ig} (e.g., the lean

and low-temperature mixtures with $Sa \approx 10$ and $\phi \approx 0.1$ as shown in Fig. 1) can still auto-ignite due to compression heating during the progress of the deflagrative burning.

2.3. Damköhler number analysis

As an alternative metric to determine the strong/weak ignition characteristics, previous studies used the Damköhler number analysis [23, 24, 27, 29, 30, 58–60], in which Da was defined as the ratio of the local reaction and diffusion terms for a selected scalar variable:

$$Da = \frac{\dot{\omega}_k}{|-\nabla \cdot (\rho Y_k \mathbf{V}_k)|}, \quad (9)$$

where Y_k , $\mathbf{V}_k$, and $\dot{\omega}_k$ denote the mass fraction, diffusion velocity, and net production rate of species k , respectively, while ρ is the mixture density. For hydrocarbon fuels, a progress of reaction variable, $Y_c \equiv Y_{CO_2} + Y_{CO}$, has been commonly used for the analysis [27–30, 59, 60].

By applying the above definition to the initial DNS solution field and quantifying Da relative to a threshold value, Da_0 , the dominant ignition regime of the mixture can be identified. Here Da_0 is of order unity and depends on the chosen initial condition. For example, it was found in a series of one-dimensional (1-D) simulations that the transition between the two propagation modes occurred approximately at $Da_0 = 4.0$ for n -C₇H₁₆/air mixtures at $p_0 = 40$ atm and $\phi_0 = 0.3$ in [27].

Base on this alternative criterion, the fractional heat release associated with strong ignition is quantified as [27–30, 59, 60]

$$F_{Da,S} = \frac{\sum \langle \dot{q} | Da > Da_0 \rangle}{\sum \dot{q}}, \quad (10)$$

where the summation is operated over the total number of computational cells in the DNS domain. Note that $F_{Da,S}$ may vary quantitatively depending on the choice of the threshold value of Da_0 , but the result was still found to be consistent [61]. In the subsequent sections, $F_{Da,S}$ will be compared with $F_{Sa,S}$ to verify the predictive accuracy of the extended ignition criterion.

2.4. Description of DNS data under study

In the subsequent analysis, previous DNS data are utilized to validate the prediction of combustion modes for different NTC fuels such as n -C₇H₁₆ [27, 30], primary reference fuels (PRF) [29, 60], and

Figure 1: (a-b) Representative initial fields of temperature and equivalence ratio (Case 16 in [30]) with $T_0 = 933$ K, $\phi_0 = 0.45$, $T' = 15$ K, and $\phi' = 0.1$, and (c-d) its corresponding ignition delay time and Sa distributions. The black iso-lines of $Sa = 1$ delineate two distinct regions of $Sa < 1$ and $Sa > 1$ which contribute to strong and weak ignition, respectively.

Figure 2: A representative probability density function of Sa distribution, $PDF(Sa)$. SI and WI denote strong and weak ignition, respectively, with a boundary of $Sa = 1$.

DME [62], as well as non-NTC fuels such as *i*-C₈H₁₈ [28] and ethanol. PRF is a blend of *iso*-octane and *n*-heptane by volume (e.g., PRF80 – a 80% *iso*-octane and 20% *n*-heptane mixture). The summary of the initial conditions for all the selected cases is described in Table 1.

The initial fields of temperature and equivalence ratio from these DNS cases were adopted to perform

the statistical analysis of Sa such that the volume-averaged (mean) Sa, $\bar{Sa}$, of the spatial Sa distribution, the probability density function of Sa, $P(Sa)$, and the volume fraction of the regions with $Sa < 1$, $F_{Sa,S}$, are obtained. In addition, Sa_p for each DNS case was also computed based on the information of the initial mean bulk mixture conditions such as T_0 , p_0 , and ϕ_0 , and the RMS values, T' and ϕ' .

The initial turbulent flow fields in all DNS cases were prescribed by an isotropic kinetic energy spectrum function [63]. Temperature and concentration fields were generated in the same manner with different random numbers to reproduce thermal and compositional non-uniformities, as shown in Fig. 1, which corresponds to a negatively correlated T - ϕ distribution with the integral length scales of temperature and equivalence ratio of $l_T = l_\phi = 1.25$ mm. All the DNS cases were performed at a constant volume with a square domain of 3.2×3.2 mm² under the homogeneous-charge compression ignition (HCCI) relevant conditions of high pressure and a wide range of the amplitude of temperature fluctuations. The integral turbulence time scale, τ_t , was chosen to be comparable to the ignition time scale, $\tau_t/\tau_{ig} \approx 1.0$, which allows significant turbulence-chemistry interactions.

Ref.	Fuel	Mechanism	T_0 (K)	p_0 (atm)	ϕ_0	T' (K)	ϕ'	l_e (mm)
Yoo et al., 2011 [27]	$n\text{-C}_7\text{H}_{16}$	58-species $n\text{-heptane}$ [27]	850, 934, 1008, 1067	40	0.3	15, 30, 60, 100	-	1.24
Yoo et al., 2013 [28]	$i\text{-C}_8\text{H}_{18}$	99-species $iso\text{-octane}$ [28]	1035	20	0.3	15, 30, 60	-	1.25
Luong et al., 2013 [29]	PRF50, PRF80, PRF100	116-species PRF [29]	1025	20	0.3	15, 30, 60	-	1.25
Luong et al., 2015 [30]	$n\text{-C}_7\text{H}_{16}$	58-species $n\text{-heptane}$ [27]	805, 933, 1025	40	0.45	15, 60	0.05, 0.1 (NC & UC)	1.25
Luong et al., 2017 [60]	PRF50	116-species PRF [29]	800, 900, 1000	40	0.45	15, 30	0.05, 0.1 (NC)	1.0
Luong et al., 2019 [62]	CH_3OCH_3	30-species DME [64]	770, 900, 1045	30	0.5	15, 30	0.075, 0.15 (NC & UC)	1.0
This study	CH_3OCH_3	30-species DME [64]	680, 770, 900, 1045	30	0.5	60	-	1.0
This study	$\text{C}_2\text{H}_5\text{OH}$	28-species ethanol [15]	880, 940, 920, 1000, 1020, 1070	40	0.5	15, 30, 60	-	1.0

Table 1: Physical parameters of all the DNS cases. Other physical parameters: Damköhler number, $\text{Da}_l = \tau_t/\tau_{ig} \approx 1.0$, the integral length scale of velocities and scalar fields of temperature and equivalence ratio, $l_e = l_T = l_\phi$. T_0 , p_0 , and ϕ_0 denote respectively the initial mean temperature, pressure, and equivalence ratio. T' and ϕ' are the root-mean-square (RMS) values of temperature and equivalence ratio, respectively. NC and UC denote negatively-correlated and uncorrelated T - ϕ distributions, respectively.

DME and $n\text{-heptane}$, exhibiting strong NTC behavior, were chosen as representative two-stage-ignition fuels to investigate the effect of temperature and equivalence ratio fluctuations on the combustion modes by varying the initial mean temperatures and different fluctuation levels [27, 30, 62]. The reduced kinetic mechanisms of 30-species for DME [64] and 58-species for $n\text{-C}_7\text{H}_{16}$ [27] were adopted. In other two DNS studies [29, 60], a 116-species reduced kinetic mechanism for primary reference fuels was used with the consideration of temperature, equivalence ratio, and reactivity fluctuations under HCCI, stratified-charge and reactivity-controlled compression ignition (SCCI, RCCI) conditions. To investigate the effect of thermal inhomogeneity under spark-assisted compression ignition (SACI) operating conditions, a 99-species reduced kinetic mechanism for $iso\text{-octane}$ was adopted [28]. An additional parametric study for a non-NTC single-stage-ignition fuel was performed by varying different initial mean T_0 and fluctuation T' of the temperature using the 28-species reduced ethanol mechanism. Six two-dimensional (2-D) DNS cases were selected where T_0 was systematically varied from 880 K to 1070 K, leading to a wide range of the homogeneous ignition delay times from 10 ms to 0.5 ms (see Fig. 3), thus covering the conditions commonly encountered in internal combustion en-

gines and shock-tube devices. Similarly, additional four 2-D cases with T_0 from 680 K to 1045 K (see Fig. 3) and T' of 60 K were performed using DME for a complete validation with the NTC two-stage-ignition fuels. Table 1 summarizes the details of the physical parameters in the DNS cases.

As a consideration of the baseline chemistry, Fig. 4 shows the homogeneous ignition delay, τ_{ig} , of $n\text{-C}_7\text{H}_{16}/\text{air}$ mixture as a function of temperature and equivalence ratio at the conditions of $p_0 = 40$ atm, $\phi_0 = 0.45$ as in [30]. To compute Sa_p , the sensitivities of ignition delay to T and ϕ , $\partial\tau_{ig}/\partial T$ and $\partial\tau_{ig}/\partial\phi$, respectively, are required. They are plotted as $\partial\log\tau_{ig}/\partial T$ and $\partial\log\tau_{ig}/\partial\phi$ in Fig. 4 and Fig. 5 for a range of conditions considered. Two main points are noted from these two figures.

First, the ignition delay time varies non-monotonically with temperature. In particular, $\partial\log\tau_{ig}/\partial T$ is negative in the low- and high-temperature regimes, whereas $\partial\log\tau_{ig}/\partial T$ is positive in the intermediate-temperature regime (i.e., from 867 K to 982 K in Fig. 4). This is referred to as the negative-temperature coefficient (NTC) regime in which the ignition delay increases with increasing temperature. Because of the non-monotonic behavior of $\partial\log\tau_{ig}/\partial T$ (distinct from the single-stage-ignition fuels), the variation of $\partial\log\tau_{ig}/\partial T$ within/near the NTC regime is shallow

Figure 3: Homogeneous ignition delay as a function of temperature for DME/air mixtures at p of 30 atm and ϕ of 0.5, and for ethanol/air mixtures at p of 40 atm and ϕ of 0.5. Selected temperatures for 2-D cases (see Table 1) are marked with symbols.

Figure 4: (a) Homogeneous ignition delay of n -heptane/air mixture as a function of temperature and its gradient with respect to temperature, $\partial \log \tau_{ig} / \partial T$, at a pressure of 40 atm, equivalence ratio of 0.45, and (b) homogeneous ignition delay as a function of equivalence ratio and its gradient with respect to equivalence ratio, $\partial \log \tau_{ig} / \partial \phi$, for three different temperatures at $p = 40$ atm.

compared to those in the low- and high-temperature regimes. These features help to explain the observed DNS results in [27, 30], where it was found

that the temperature fluctuation is less effective in reducing the pressure rise rate if T_0 is within/near the NTC regime. Therefore, it is expected that the mixture within the NTC is more susceptible to autoignition if no concentration fluctuations exist. This behavior on the ignition mode is further quantified by $\text{PDF}(\text{Sa})$, $F_{\text{Sa},S}$, $\bar{\text{Sa}}$, and Sa_p in the following sections under various conditions.

Second, in contrast to the non-monotonic behavior of $\partial \log \tau_{ig} / \partial T$, $\partial \log \tau_{ig} / \partial \phi$ monotonically decreases with increasing ϕ . The resultant effect of the T and ϕ variations on τ_{ig} is shown in Fig. 5, along with the behavior of its derivatives with respect to T and ϕ . It is seen that the NTC characteristics ($\partial \log \tau_{ig} / \partial T > 0$) are mainly due to the temperature effects. In the previous DNS studies [25, 30, 59, 60, 65, 66], a negatively-correlated T - ϕ distribution, representing the evaporative cooling effect of fuels, suppresses the deflagration development if T_0 is outside the NTC regime, while it has a synergistic effect if T_0 is inside the NTC regime. As such, the extended predictive criterion of the combustion modes following Eq. 7 in the presence of both T' and ϕ' at different bulk mixture conditions needs to be examined.

3. Results and discussion

Based on the 0-D ignition data shown in Fig. 5, the overall ignition characteristics in terms of weak versus strong ignition are predicted. The roadmap of the analysis is as follows. First, for different DNS results the PDF and volume-averaged quantities of the exact Sa are computed and their correlation with the fractional heat release, $F_{\text{Da},S}$, is assessed. It will be demonstrated that the local evaluation of exact Sa according to Eqs. 5 and 7 correctly predicts the heat release behavior for all cases under consideration. Therefore, the first point that the Sankaran criterion can be extended to a general mixture temperature and composition fluctuations will be made. Subsequently, considering that the exact Sa is unavailable in general engineering Reynolds-averaged Navier-Stokes (RANS) simulations, the predicted Sa , Sa_p , is computed by using Eq. 5, Eq. 7, and the mean quantities, T_0 , ϕ_0 and p_0 , for all DNS data and its accuracy in predicting the same ignition metric is assessed. As such, it will be shown that the Sa_p criterion can be used to predict the ignition characteristics of nearly homogeneous mixtures with temperature and composition fluctuations, including NTC fuels.

Figure 5: (a) The homogeneous ignition delay of *n*-heptane/air mixtures in the T - ϕ space at $p = 40$ atm, and its derivative with respect to (b) temperature, $\partial \log \tau_{ig} / \partial T$, and (c) equivalence ratio, $\partial \log \tau_{ig} / \partial \phi$. The boundary iso-line marked by 0.0 delineates the NTC regime in the T - ϕ space.

For a systematic progression of discussion, the overall combustion characteristics of the ethanol/air mixtures and the DME/air mixtures are discussed first, followed by the analysis of deterministic prediction of different criteria.

3.1. Overall combustion characteristics

To have a comprehensive picture of validation, in addition to the previous DNS studies, ten additional cases covering a wide range of ignition delay times are performed by varying T_0 , T' , and the T' - ϕ' combination. The temporal evolution of mean HRR and iso-contours of HRR for some of these cases are shown in Fig. 6 and Fig. 7 for ethanol, and in Fig. 8 and Fig. 10 for DME.

3.1.1. Temperature inhomogeneities

In general, decreasing the initial mean temperature and/or increasing the fluctuation levels promote deflagration fronts, prolonging the combustion duration and lowering the maximum HRR. As such, a weak combustion mode is observed for all the cases of ethanol shown in Fig. 6 and Fig. 7, even for the case with T_0 of 900 K and T' of 15 K. Regardless of the variation in T' and T_0 , the maximum mean HRR for these three cases is of the same order of magnitude as shown in Fig. 6b. Note that these three cases were selected such that they have nearly identical $F_{Sa,S}$ of 0.3.

In contrast to the weak-ignition cases of ethanol, spontaneous ignition (characterized by high peak HRR in Fig. 8 and Fig. 10) is dominant even with T' of 60 K for the DME case at T_0 of 900 K due to a small variation of τ_{ig} with temperature within/near the NTC regime as shown in Fig. 3), whereas flame propagation is dominant for the cases with T_0 beyond the NTC regime (i.e., T_0 of 680 K and 1045

Figure 6: Temporal evolution of the mean HRR for ethanol/air mixtures at p_0 of 40 atm and ϕ_0 of 0.5 with (a) T' of 60 K and varying T_0 , (b) nearly identical $F_{Sa,S}$ of 0.3, and varying both T' & T_0 . The HRR contour of each case is normalized by its corresponding 0-D maximum HRR.

K) for the same level of temperature fluctuation, T' of 60 K. Similar results were found for *n*-heptane [27, 30] (discussed in Section 3.2.1), and PRF50 and PRF80 [67].

As a further quantitative verification, $\bar{S}_a$ and $F_{Sa,S}$ for the DME cases of T_0 at 680 K, 900 K, and

Figure 7: 2-D iso-contours of the normalized HRR at τ_{ig} at which the maximum spatial-mean HRR occurs for the ethanol/air mixtures. The top row corresponds to the cases with T' of 60 K, and the bottom row corresponds to the three cases with nearly identical $F_{Sa,S}$ of 0.3, but with different T' . For a direct comparison, the HRR contour of each case is normalized by its corresponding 0-D maximum HRR.

Figure 8: Temporal evolution of the mean heat release rate for DME/air mixtures at p_0 of 30 atm and ϕ_0 of 0.5. The HRR contour of each case is normalized by its corresponding 0-D maximum HRR.

1045 K are found to be, respectively, 4.2 & 0.20, 0.5 & 0.92, and 1.3 & 0.4. The cases with T_0 of 680 K and 1045 K have $\bar{S}_a$ greater than one, suggesting a weak-ignition dominance, while for the case with T_0 of 900 K, a strong ignition is expected. The predictive accuracy of $\bar{S}_a$ and $F_{Sa,S}$ is further verified

by the magnitude of $F_{Da,S}$ that will be discussed in the next section.

3.1.2. Temperature and concentration inhomogeneities

The temporal evolution of HRR profiles for DME cases from [62] with T' of 30 K and ϕ' is shown in Fig. 9 at three distinct low-, intermediate-, and high-temperature regimes. The HRR contours of three representative cases of $\phi = 0.15$ are shown at the bottom row of Fig. 10. Contrary to the T' -only cases, in the presence of both T and ϕ inhomogeneities and the negative T - ϕ correlation, T_0 of 900 K results in a much lower HRR (due to the synergistic effect of T' and ϕ') than those of T_0 outside the NTC regime (due to the subtractive effect of T' and ϕ' for the cases with T_0 of 770 K and 1045 K), as seen in the bottom row of Fig. 10. Quantitatively, $\bar{S}_a$ and $F_{Sa,S}$ for the three cases in Fig. 10 are 0.5 & 0.85, 2.1 & 0.56, and 0.3 & 0.95 for T_0 of 770 K, 900 K, and 1045 K, respectively. The prediction of $F_{Sa,S}$ is in agreement with the HRR contours regardless of the presence of the T - ϕ inhomogeneities.

These observations can be explained by Eq. 7.

Figure 9: Temporal evolution of the mean heat release rate for DME/air mixtures with $T-\phi$ fluctuations at T_0 of 770 K, 900 K, and 1045 K, p_0 of 30 atm, and ϕ_0 of 0.5. NC and UC denote the negative correlation and uncorrelated $T-\phi$ distribution.

Figure 10: 2-D iso-contours of the normalized HRR at τ_{ig} at which the maximum spatial-mean HRR occurs for the DME/air mixtures. The top row corresponds to the cases with temperature fluctuations only, T' of 60 K, and the bottom row corresponds to the cases with both temperature and equivalence ratio fluctuations, T' of 30 K and ϕ of 0.15 [62]. For a direct comparison, the HRR contour of each case is normalized by its corresponding 0-D maximum HRR.

According to Eq. 7, the contributions of temperature and equivalence ratio to Sa_p can be discerned by examining $T' \partial \tau_{ig} / \partial T$ and $\phi' \partial \tau_{ig} / \partial \phi$. Table 2 lists the values of these terms for six DME cases with a negatively-correlated $T-\phi$ distribution [62].

As readily seen in Table 2, the absolute magnitudes of $T' \partial \tau_{ig} / \partial T$ and $\phi' \partial \tau_{ig} / \partial \phi$ are comparable, having contributions to Sa_p of the same order. If they are subtractive (i.e. T_0 of 770 K and 1045 K with negatively-correlated $T-\phi$), the resulting HRR pro-

Case	T_0 (K)	T' (K)	ϕ' -	$T' \partial \tau_{ig} / \partial T$ (ms $\times 10^{-2}$)	$\phi' \partial \tau_{ig} / \partial \phi$ (ms $\times 10^{-2}$)	$\bar{S}a$	Sa_p
<i>n</i>-Heptane							
1	805	15	0.05	-21.5	-23.4	0.11	0.09
2	805	15	-	-21.5	-	0.21	0.42
3	805	-	0.05	-	-23.4	0.19	0.4
4	933	15	0.05	15.6	-19.6	0.57	1.40
5	933	15	-	15.6	-	0.26	0.55
6	933	-	0.05	-	-19.6	0.27	0.62
7	1025	15	0.10	-24.6	-27.4	0.14	0.16
8	1025	15	-	-24.6	-	0.65	1.23
9	1025	-	0.10	-	-27.4	0.55	1.08
DME							
1	770	15	0.075	-13.1	-8.6	0.2	0.1
2	770	30	0.150	-26.2	-17.2	0.5	0.3
3	900	15	0.075	5.2	-13.6	0.5	1.0
4	900	30	0.150	10.4	-27.2	2.1	2.0
5	1045	15	0.075	-11.6	-8.8	0.1	0.2
6	1045	30	0.150	-23.2	-17.6	0.3	0.4

Table 2: The mean and predicted Sankaran numbers, $\bar{S}a$ and Sa_p , along with the contributions from the temperature and equivalence ratio terms, for nine representative 2-D *n*-heptane cases [30] with either T' or ϕ' only and a negatively correlated (NC) T - ϕ distribution, and for six 2-D DME cases [62] with a NC T - ϕ distribution.

files are similar to 0-D ignition (spontaneous ignition), which is reflected in a high peak HRR (see Fig. 9). On the other hand, if they are additive (T_0 of 900 K), the peak of HRR is reduced. Note also that for the case T_0 of 900 K inside the NTC regime, both negative and uncorrelated T - ϕ distributions result in an additive effect on Sa_p such that with the same level of T' and ϕ' , nearly identical HRR profiles are obtained, except for the difference in the ignition delay time (see Fig. 9).

Spontaneous ignitions were also found for *n*-heptane with T_0 of 805 K and 1025 K (see Fig. 6 in [30]) due to the offsetting effect of the comparable $T' \partial \tau_{ig} / \partial T$ and $\phi' \partial \tau_{ig} / \partial \phi$ terms (Cases 1 and 7 in Table 2). In line with Eq. 7, Table 2 also allows to verify that at the same T_0 , the cases with either T' or ϕ' only have nearly identical $T' \partial \tau_{ig} / \partial T$ and $\phi' \partial \tau_{ig} / \partial \phi$ (i.e., T_0 of 805 K with either T' of 15 K or ϕ' of 0.05, and T_0 of 1025 K with either T' of 15 K or ϕ' of 1.0), which in turn have comparable $\bar{S}a$ and share similar combustion characteristics (see Fig. 6 in [30]).

Consistent findings were reported with different T_0 and different fuels such as *n*-heptane [30, 65, 66], PRF50 [60], biodiesel [59], hydrogen [25]. In a recent study [57], these criteria were used to predict the combustion intensity using LES initial conditions generated from a full cycle engine simulation

under direct-injection SI engine conditions over a wide temperature range from 700 K to 1280 K. The prediction was compared with RANS-based simulation data and the results were found to be consistent with the present findings, namely $\bar{S}a$ and Sa_p show good agreement, and the correlation of Sa_p with the combustion intensity is consistent with Fig. 14 [57]. The results from the DNS studies, and the Sa -based prediction for the DME and *n*-heptane cases are also qualitatively consistent with previous experimental findings. The experimental studies in References [68, 69] found that inappropriate mixture preparation by the fuel-direct injection can cause an adverse effect on HCCI combustion, leading to excessive HRR due to the offset effect of temperature and fuel inhomogeneities. Dec and co-authors [3, 11, 70] found that fuel stratification reduces the maximum PRR under highly boosted conditions, which is attributed to the NTC-chemistry enhancement of *iso*-octane-like fuels such as gasoline; however, no benefit of the maximum PRR reduction was achieved under naturally aspirated intake conditions.

3.2. Quantitative prediction of combustion modes

As discussed in the previous section, the combustion modes are highly sensitive to the initial mean temperatures, fuel types, and temperature and con-

centration fluctuations. A robust metric to quantify the combustion mode, combustion intensity, and ignition delay times is needed. In this regard, different predictive criteria and their predictive accuracy will be examined in the following section.

3.2.1. Probability density function of Sa

For the n -heptane/air mixture cases with temperature fluctuations only [27], the initial solution fields were processed and the PDF of Sa distribution is shown in Fig. 11. For most cases, Sa distribution is mainly within the range of $Sa < 1$, suggesting that they are in the strong ignition regime. In general, for a given T' , a higher bulk temperature shifts the distribution further towards a lower Sa , and for a given T_0 , a higher T' widens the distribution. However, the NTC characteristics are also manifested in that the Sa distribution is distinctly shifted to lower values for $T_0 = 850$ and 934 K. As such, a mixture with a T_0 outside the NTC range with a higher T' is more likely to develop weak ignition behavior. Indeed, Yoo et al. [27] reported that the resulting heat release behavior was found to be as expected from the Sa distribution. Additional simulation data by Luong et al. [30] for n -heptane/air mixtures at different ϕ_0 and T_0 conditions also showed consistent PDF behavior (not shown here).

3.2.2. Comparison between $F_{Sa,S}$ and $F_{Da,S}$

As a further quantitative assessment, Fig. 12 shows the actual computed values of the fraction of heat release due to strong ignition, $F_{Da,S}$, plotted against the corresponding volumetric fraction of $Sa < 1$, $F_{Sa,S}$, based on the PDF statistics such as those in Fig. 11. For all the cases considered, $F_{Da,S}$ and $F_{Sa,S}$ are found to correlate very well, even for the cases in which T' is as large as 100 K [27] and even though the bulk temperature of some cases falls into the NTC range. There appears to be a systematic shift in the correlation for ethanol and DME cases, which may need to be calibrated further. Nevertheless, all correlations follow approximately a straight line, suggesting that $F_{Sa,S}$ serves as a good predictive marker to characterize the overall ignition regimes.

3.2.3. Statistical mean Sankaran number, $\bar{Sa}$

Next, it is of interest to assess an alternative metric of the volume-averaged Sankaran number, $\bar{Sa}$, in its prediction of the ignition behavior. Fig. 13 shows the correlation between $F_{Sa,S}$ and $\bar{Sa}$ and

Figure 11: The probability density function (PDF) of Sa , $P(Sa)$, at different initial mean temperatures with (a) $T' = 15$ K, (b) $T' = 30$ K, and (c) $T' = 60$ K.

Sa_p . For both Sa numbers, the subscript n indicates that the quantities are re-scaled as $Sa/(1 + Sa)$ to fit within a range between 0 and 1, with 0.5 corresponding to $Sa = 1$. All DNS cases are compiled and organized for different fuels. A strong correlation between $\bar{Sa}_n$ and $F_{Sa,S}$ is observed regardless of T_0 , T' , and $T-\phi$. The correlation is fairly good not only for n -heptane, DME (also in Fig. 4d in [62]), and a single-stage ignition fuel, ethanol, but it is also observed in other fuel/air mixtures under various conditions such as PRF50 and *iso*-octane (not shown here).

Figure 12: The HRR fraction of spontaneous ignition (SI), $F_{Da,S}$, obtained by a Damköhler-number-based analysis in the previous DNS studies [27–30, 60, 62] and the present study (ethanol) as a function of the fraction of $Sa < 1$, $F_{Sa,S}$. The solid line denotes the ratio of $F_{Da,S}/F_{Sa,S} = 1$. The filled symbols corresponding to the T' -only cases and the open symbols corresponding to the $T' - \phi'$ cases are consistently used in Figs. 12–16.

Figure 13: The volumetric fraction of $Sa < 1$, $F_{Sa,S}$, as a function of (a) the normalized $\bar{Sa}$, $\bar{Sa}_n$, and (b) the normalized Sa_p , $Sa_{p,n}$.

Figure 14: Comparison of the normalized mean $\bar{Sa}$ and normalized predicted Sa , $\bar{Sa}_n$ versus $Sa_{p,n}$.

Based on the strong correlations shown in Fig. 13, the magnitude of $\bar{Sa}$ can be used to directly predict $F_{Sa,S}$. In general, a higher $\bar{Sa}$ corresponds to a lower $F_{Sa,S}$, which translates into a lower combustion intensity. In particular, $F_{Sa,S} = 0.6$ approximately corresponds to $\bar{Sa} = 1.0$ ($\bar{Sa}_n = 0.5$) as a transition point from SI to WI. At this condition, the fractional contribution of the heat release by spontaneous ignition is also about 60%. For $\bar{Sa} < 0.3$, $F_{Sa,S} = 1.0$ corresponds to purely spontaneous ignition. On the contrary, for $\bar{Sa} > 1.0$, weak ignition is predicted to be predominant. Therefore, it is demonstrated that $\bar{Sa}$ can be used to quantitatively predict the combustion modes.

As for the second correlation in Fig. 13(b), despite some scattering, the $Sa_{p,n}$ - $F_{Sa,S}$ correlation appears to be also well reproduced, especially in the range of both strong and weak ignition limits, say $F_{Sa,S} < 0.2$ and $F_{Sa,S} > 0.8$. This suggests that $Sa_{p,n}$ can also capture the strong/weak ignition behavior, although at some loss of accuracy in the mixed mode of strong/weak ignition regimes.

3.2.4. Comparison between $\bar{Sa}_n$ and $Sa_{p,n}$

The normalized predicted $Sa_{p,n}$ is validated by comparing it with the normalized statistical mean $\bar{Sa}_n$ for four different datasets with and without the NTC behavior as shown in Fig. 14. Overall, $Sa_{p,n}$ exhibits a good agreement with $\bar{Sa}_n$ in spite of the discrepancy that shows $Sa_{p,n} > \bar{Sa}_n$ for the most points considered. In general, lowering initial mean temperatures and/or increasing the fluctuation levels (i.e., increased T' and/or ϕ') result in a wider distribution of Sa (e.g., see PDF(Sa) in Fig. 11), thereby increasing $\bar{Sa}_n$ significantly because τ_{ig} in-

Figure 15: The normalized maximum spatial-mean HRR, $\bar{q}_m/\dot{q}_m^0$, as a function of (a) the normalized $\bar{S}_a$, $\bar{S}_{a,n} = \bar{S}_a/(\bar{S}_a+1)$, (b) the normalized $S_{a,p}$, $S_{a,p,n} = S_{a,p}/(S_{a,p}+1)$, (c) $F_{S_{a,S}}$ and (d) $F_{D_{a,S}}$. The solid line shows the fitting correlation $\bar{q}_m/\dot{q}_m^0$ as a function of $\bar{S}_{a,n}$, $f(\bar{S}_{a,n}) = 0.945 \exp(-3.187\bar{S}_{a,n})$.

creases exponentially with decreasing temperature. Therefore, for the cases with a wide τ_{ig} distribution, $\bar{S}_a$ is computed by excluding the grid points with $\tau_{ig} > 2\tau_{ig}^0$ (τ_{ig}^0 is computed at T_0 , ϕ_0 and p_0) in order to obtain a well-correlated pattern of $\bar{S}_{a,n}$ and $F_{S_{a,S}}$; otherwise, there will be large discrepancies between $S_{a,p,n}$ and $\bar{S}_{a,n}$ leading to degraded correlations in the $\bar{S}_{a,n}-F_{S_{a,S}}$ distribution. Excluding the grid points with $\tau_{ig} > 2\tau_{ig}^0$ in computing $\bar{S}_a$ is justifiable because these locations are of little significance in determining the ignition behavior as the onset of combustion is driven by the most reactive spots that have the shortest ignition delays.

3.2.5. Correlation between the maximum HRR and $\bar{S}_{a,n}$, $S_{a,p,n}$

As an additional relevant physical observable in the ignition system, quantitative prediction of the combustion intensity is further demonstrated by the ratio of $\bar{q}_m$ and $\dot{q}_m^0$. The maximum spatial-mean HRR, $\bar{q}_m$, normalized by its corresponding maximum 0-D homogeneous HRR, $\dot{q}_m^0$, is plotted as a function of $\bar{S}_{a,n}$ and $S_{a,p,n}$ in Fig. 15. A good cor-

relation between $\bar{q}_m$ and $\bar{S}_{a,n}$ is observed in Figure 15a, and a consistent correlation between $\bar{q}_m/\dot{q}_m^0 - S_{a,p,n}$ is also found in Fig. 15b.

The ratio $\bar{q}_m/\dot{q}_m^0$ is approximately equal to 0.2 for $\bar{S}_a$ of unity. As $\bar{S}_a$ takes on values much smaller than unity, $\bar{q}_m/\dot{q}_m^0$ increases exponentially and approaches unity, such that the purely spontaneous ignition mode becomes dominant. The correlation between $\bar{q}_m$ and $\bar{S}_{a,n}$ is fitted well by an exponential function as $f(\bar{S}_{a,n}) = 0.945 \exp(-3.187\bar{S}_{a,n})$ (the thick line in Fig. 15a).

The scatter plot of $\bar{q}_m/\dot{q}_m^0$ and $F_{S_{a,S}}$ ($F_{D_{a,S}}$) also shows in Fig. 15c-d a similar monotonic and exponential increase in $\bar{q}_m/\dot{q}_m^0$ with $F_{S_{a,S}}$ ($F_{D_{a,S}}$). Fig. 15 also suggests that $\bar{S}_{a,n}$ and $S_{a,p,n}$ are better metrics to correlate with $\bar{q}_m/\dot{q}_m^0$ because the variation in $\bar{q}_m/\dot{q}_m^0$ is not reflected by $F_{S_{a,S}}$ and $F_{D_{a,S}}$ when $F_{S_{a,S}}$ and $F_{D_{a,S}}$ approach unity, while $\bar{S}_{a,n}$ and $S_{a,p,n}$ monotonically decrease with increasing $\bar{q}_m/\dot{q}_m^0$.

Figure 16: The normalized ignition delay, τ_{ig}/τ_{ig}^0 , as a function of (a) the normalized $\bar{S}a_n$, $\bar{S}a_n = \bar{S}a/(\bar{S}a+1)$, (b) the normalized $S_{a,p}$, $S_{a,p,n} = S_{a,p}/(S_{a,p}+1)$. The solid line shows their polynomial fitting correlation.

3.2.6. Correlation between the ignition delay time and $\bar{S}a_n$, $S_{a,p,n}$

Finally, the ignition delay times plotted as a function of $\bar{S}a_n$ and $S_{a,p,n}$ are examined in Fig. 16. A good correlation between τ_{ig}/τ_{ig}^0 and $S_{a,p,n}$ is found for the cases of the single-stage-ignition fuel (i.e., ethanol) and for the cases with T_0 lying outside the NTC regime (e.g., DME with T_0 of 680 K) because τ_{ig}/τ_{ig}^0 decreases monotonically with increasing T' and/or ϕ' . It is also seen that τ_{ig}/τ_{ig}^0 exponentially decreases with increasing $S_{a,p,n}$ for $S_{a,p,n} > 0.5$ ($S_{a,p} > 1$). However, previous studies reported that the overall combustion was delayed (i.e., τ_{ig} slightly increased, $\tau_{ig}/\tau_{ig}^0 > 1.0$) if T_0 approached the NTC regime [27, 30, 59]; hence, the widely-scattered distribution of τ_{ig}/τ_{ig}^0 - $\bar{S}a_n$ ($S_{a,p,n}$) in Fig. 16 is expected for those cases due to the NTC behavior. The normalized ignition delay time of these cases is close to 1.

Similar findings were reported in recent shock

tube studies [71, 72]. Generally, if a mixture in the shock tube experienced preignition caused by hot spots, its ignition delay time was found to be much shorter than those of 0-D reactors [71, 72]; however, for the cases of *n*-heptane with T_0 lying in the intermediate-to-low-temperature regime, τ_{ig}/τ_{ig}^0 of the bulk mixture was found to be greater than unity [71].

4. Conclusions

The ignition regime criteria proposed by Im et al. [1] was extended to consider both temperature and concentration fluctuations. The auto-ignition modes of different fuels with and without the NTC regime together with thermal and/or compositional non-uniformities were examined to validate the extended criteria.

Extensive DNS results were used to validate the predictive accuracy of the proposed criteria. It was demonstrated that the combustion modes of strong/weak ignition were well captured by the normalized statistical mean Sankaran number, $\bar{S}a_n$ and the volumetric fraction of $Sa < 1$, $F_{Sa,S}$. Moreover, $\bar{S}a_n = 0.5$ ($\bar{S}a = 1$) was found as a predictive metric to identify the transition between the weak and strong ignition regimes.

In addition, the HRR fraction from strong ignition can be quantified by $F_{Sa,S}$. $F_{Sa,S}$ shows a satisfactory agreement with $F_{Da,S}$ from the DNS analysis. $F_{Sa,S}$ can also be approximated by the relative magnitude of $\bar{S}a_n$. At $\bar{S}a_n = 0.5$, the $F_{Sa,S}$ is approximately equal to 0.6, suggesting that 60% (40%) heat release attributed from spontaneous ignition (deflagration). As $\bar{S}a$ increases turning greater than unity, higher $S_{a,p}$ yields more deflagration fronts, ultimately resulting in the dominant deflagration mode (weak ignition).

Finally, the predicted Sa , $S_{a,p}$, is in good agreement with $\bar{S}a$. It was also found that both $\bar{S}a$ and $S_{a,p}$ correlate very well with the normalized maximum heat release. Moreover, a good correlation between the ignition delay time and $S_{a,p}$ is observed for single-stage fuels, and NTC fuels if T_0 lies outside the NTC regime. In summary, the validity of the ignition regime criteria [1] considering temperature/composition inhomogeneities has been demonstrated for a variety of fuels.

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