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Ignition Process in a Non-Homogeneous Mixture

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1. Introduction

An auto-ignition process of a non-homogeneous mixture is investigated using a numerical calculation based on chemical kinetics and the stochastic approach. This type of auto-ignition phenomenon is considered as a fundamental process of the initial stage of diesel combustion (Ishiyama, et al., 2003) or homogeneous charged compression ignition (Shimasaki, et al., 2003). In order to investigate these combustion processes, many numerical calculations have been performed and for many of those it has been assumed that the ignition process is dominated by the turbulent mixing (Kong and Reitz, 2000).

However, the fuel-air mixing and chemical reaction progress happen simultaneously for these types of combustion processes. Due to the long ignition delay time and high homogeneity of the mixture, combustion characteristics, such as ignition delay and combustion duration, could be affected equally by non-homogeneity of the mixture, turbulent mixing rate and chemical reaction rate. Therefore, the understanding of those combustion mechanisms is incomplete due to the complexity of the phenomena in which the mixing and chemical processes interact with each other. The main purpose of this chapter is to estimate these effects quantitatively using a numerical method.

Here, n-heptane is assumed as a fuel and its reaction process is calculated by means of a reduced mechanism (Seiser et al., 2000). The non-uniform states of turbulent mixing are statistically described using probability density functions and the stochastic method, which was newly developed from Curl's model (Curl, 1963). Focusing on the effects of mixture heterogeneity on combustion characteristics, such as ignition delay and combustion intensity (rate of temperature rise), the evolution of chemical reactions was calculated for the mixture in which variance in fuel-mass fraction decreases at given rates from the initial value under a fixed mean fuel-mass fraction and a constant pressure. The results show that the start timing of the low-temperature oxidation and ignition delay period are hardly affected by the equivalence-ratio variation, however, combustion duration increases with increasing variance. Furthermore, the combustion duration is mainly affected by the non-homogeneity at the ignition and is not much affected by the mixing rate.

2. Numerical models

2.1 Mixing model

In order to describe the homogenization process of a fuel-air mixture by turbulent mixing, the statistical state of the mixture is expressed by means of probability density function (PDF) and the dissipation process is described by a particle-interaction model. In the particle-interaction model, the statistical state f of scalar ϕ at time t is represented by the N delta functions δ;

$$f(\phi;t) = \frac{1}{N} \sum_{n=1}^{N} \delta(\phi - \phi(n)) \tag{1}$$

Here, $\phi*(n)$ indicates ϕ value on the nth particle. In the coalescence/dispersion model proposed by Curl (1963), the state of f at $t + \Delta t$ is calculated from the collision frequency ω. The compositions of a pair of particles, which are selected at random (denoted by n_1 and n_2), change as;

$$\phi*(n_1)\big|_{t=t+\Delta t} = \phi*(n_2)\big|_{t=t+\Delta t} = \frac{1}{2}\left(\phi*(n_1)\big|_{t=t} + \phi*(n_2)\big|_{t=t}\right) \tag{2}$$

Then, within the time interval Δt, this operation is repeated $\Delta t N \omega$ times.

The change in the value of $\phi*$ is sometimes large, especially in the early stages of mixing, because it is calculated as the average of significantly different quantities of two particles. This tends to cause unrealistic change in the progress of chemical reactions. The modified Curl's model (Janicka, Kolbe and Kollmann, 1979) is one possible method to mitigate this tendency. In this method, the scalar exchange of a pair of particles (n_1 and n_2) is calculated from a uniform random number C between 0 and 1;

$$\phi*(n_1)\big|_{t=t+\Delta t} = \phi*(n_1)\big|_t + \frac{1}{2}C\left(\phi*(n_2)\big|_{t=t} - \phi*(n_1)\big|_{t=t}\right)$$
$$\phi*(n_2)\big|_{t=t+\Delta t} = \phi*(n_2)\big|_t - \frac{1}{2}C\left(\phi*(n_2)\big|_{t=t} - \phi*(n_1)\big|_{t=t}\right) \tag{3}$$

This operation is repeated $3/2\Delta t N \omega$ times and can describe statistically the same process as Curl's model. However, the value of $\phi*_{t+\Delta t}$ still changes significantly if C is near 1. Here, in order to avoid a significant difference between $\phi*_t$ and $\phi*_{t+\Delta t}$, C is fixed to a value sufficiently smaller than unity and the replacement procedures are repeated $\Delta t N \omega /(2C)$ times. In order to confirm the consistency between the present model and Curl's model, a mixing process was calculated starting from the initial state in which fuel and air are perfectly separated.

Time change of variance v_m for fuel-mass fraction mf distribution is shown in Fig. 1. Here, v_m is normalized with respect to initial variance v_{mi} and dimensionless time $t* = \omega t$ is used. $C = 0.05$ and $N = 1000$ are adopted. As shown, the time changes of v_m/v_{mi} calculated by these three methods are completely equivalent. Meanwhile, Figure 2 shows the time change in m_f-PDF. The distributions at smaller t calculated by the present model are similar to Gaussian rather than to the PDFs by the Curl's and modified Curl's models.
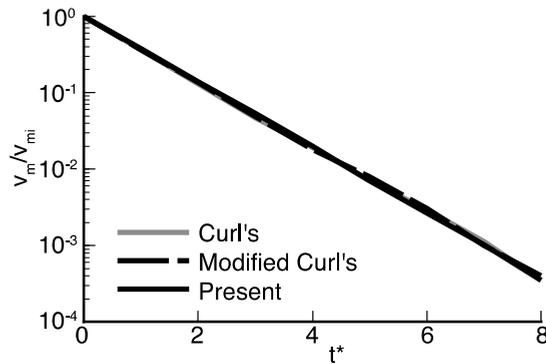
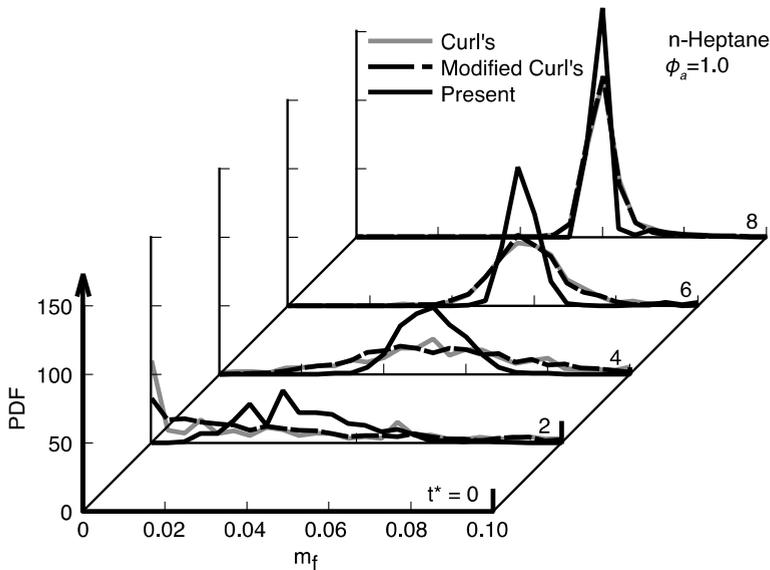Fig. 1. Course of normalized variance v_m/v_{mi} against non-dimensional time t^*

Fig. 2. Temporal change in PDF of fuel-mass fraction m_f

2.2 Chemical reaction model

Here, n-heptane is assumed as a fuel and a chemical reaction system is described by a semi-detailed kinetic mechanism (Seiser et al., 2008[Web]). This system consists of 160 chemical species and 1540 elemental reactions, and is selected for reducing computation time.

3. Results and discussion

By means of the above-mentioned procedure, an ignition process of non-homogeneous mixture with a constant mixing rate is calculated. For simplicity, the calculation is performed under a constant pressure. The initial temperature T_i and pressure p are set to

T_i = 900K and p = 4MPa. The initial distribution of fuel-mass fraction is Gaussian with a mean value m_{fa} and a variance v_{mi}. Figure 3 shows time histories of mean temperature T_a and variance v_m for the initial condition of m_f = 6.22×10⁻² and v_{mi} = 4.34×10⁻⁵, corresponding to a mean equivalence ratio ϕ_a of unity and the standard deviation of ϕ of approximately 0.1. The collision frequency ω is varied within the range of 0 s⁻¹ to 10000 s⁻¹. For comparison, the result for the case of the homogeneous mixture is also shown. Collision frequency ω is usually given by $\omega = C_m \varepsilon / k$ for the PDF model combined with CFD using the k-ε turbulence model.
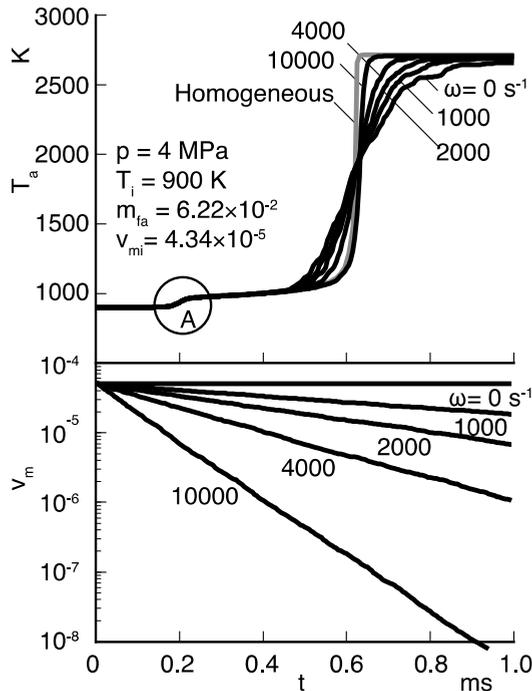

Fig. 3. Courses of mean-temperature T_a and v_m for the non-homogeneous mixture (ϕ_a = 1.0)

Here, k and ε represent turbulence energy and its dissipation rate, respectively. C_m is fixed at 2, therefore, ω = 2000 s⁻¹ corresponds approximately to turbulence intensity of 5 m/s and scale of 3 mm. For these calculations, the number of fluid particles is fixed to N = 100. Generally, as collision frequency increases, the start of heat release by hot flame delays and maximum heat release rate increases simultaneously to approach the result of a homogeneous mixture. Meanwhile, the time when the temperature reaches 50% of adiabatic flame temperature is approximately constant regardless of the collision frequency ω. The reason will be discussed later in Fig. 8. Furthermore, Fig. 4 shows a magnified view of Fig. 3 around the starting points of heat release by cool flame (marked as 'A'). The change in mean temperature T_a is exactly the same, in spite of the change of ω.

Next, in order to clarify the effect of mean equivalence ratio, calculations are performed for lean and rich cases. Fig. 5 shows T_a and v_m under the conditions of (a) m_{fa} = 3.87×10⁻² (ϕ_a = 0.6) and (b) m_{fa} = 1.17×10⁻¹ (ϕ_a = 2.0) for the same ω as in Fig. 3. Here, initial value of variance
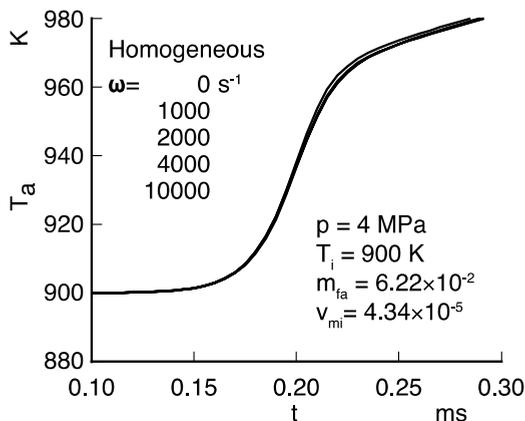
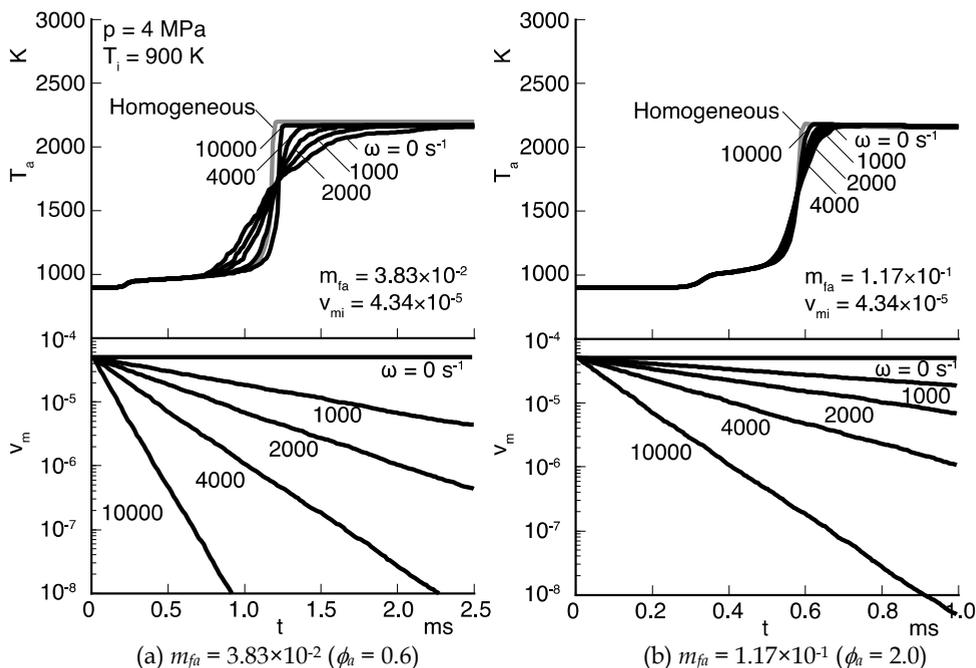Fig. 4. Temporal changes of Ta due to low temperature oxidation

Fig. 5. Effect of mean mass-fraction m_{fa}

is fixed to $v_{mi} = 4.34 \times 10^{-5}$. For the two conditions, histories of the mean temperature T_a are similar to the result of $\phi_a = 1.0$ in Fig. 3. For the lean case (a), due to the longer delay of hot flame heat release, the mixture is thought to be more homogeneous and to provide rapid temperature rise. However, the temperature-rising rate at ignition is lower compared to the result of $\phi_a = 1$. Meanwhile, for the rich case (b), the variance v_m at the ignition is comparably large, whereas the temperature rise at the ignition is steep, similar to the homogeneous case.

In order to clarify the reason why the temperature-rising rate of the non-homogeneous mixture becomes smaller than in the homogenous case, the temporal change of temperature T and mass fraction of fuel mf in each fluid particle are examined. Fig. 6 shows the data of
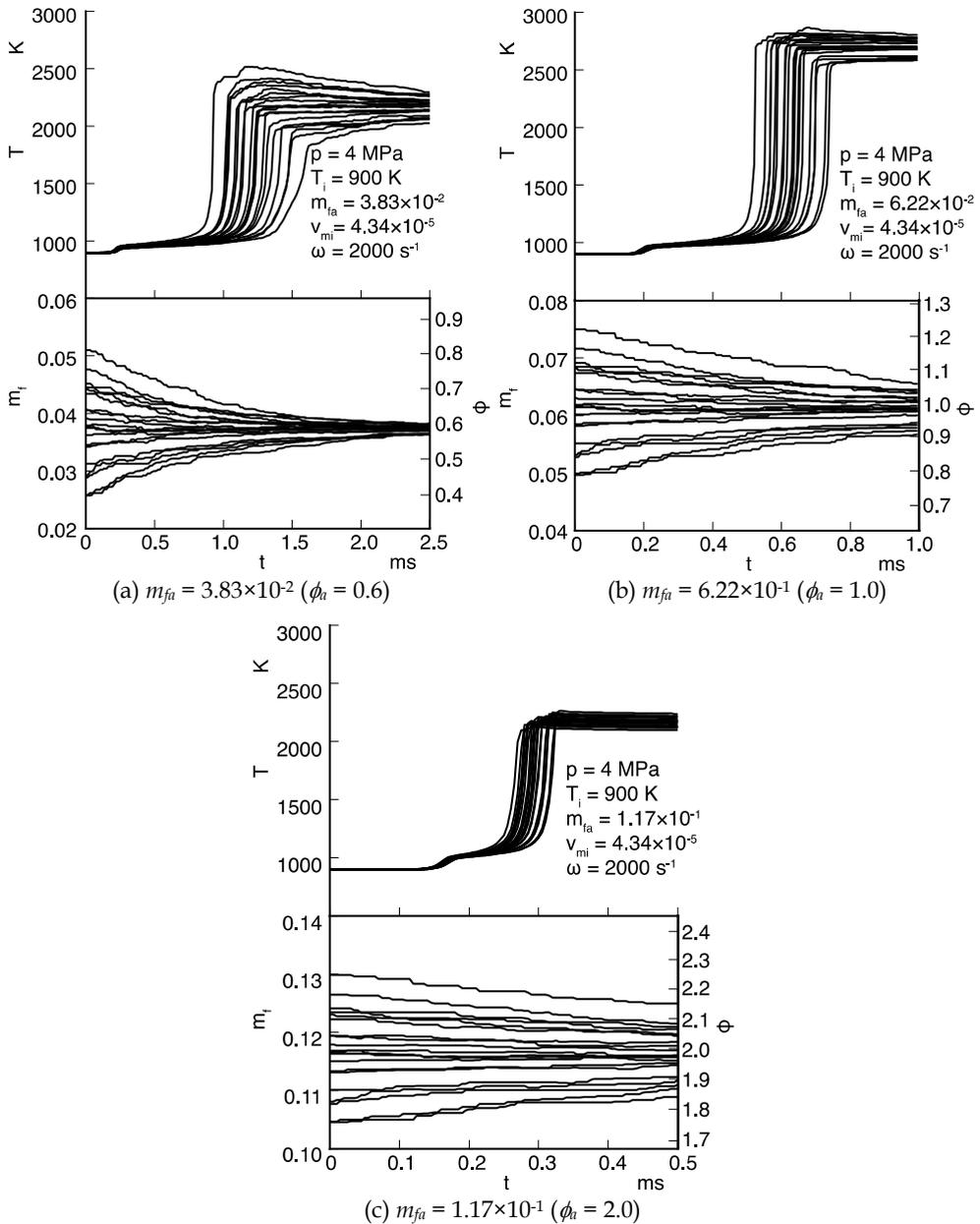

(a) $m_{fa} = 3.83 \times 10^{-2}$ ($\phi_a = 0.6$)

(b) $m_{fa} = 6.22 \times 10^{-1}$ ($\phi_a = 1.0$)

(c) $m_{fa} = 1.17 \times 10^{-1}$ ($\phi_a = 2.0$)

Fig. 6. Time history of T and m_f for each stochastic particle

20 fluid particles, which are selected randomly, for (a) m_{fa} = 3.83×10⁻² (ϕ_a = 0.6), (b) m_{fa} = 6.22×10⁻² (ϕ_a = 1.0) and (c) m_{fa} = 1.17×10⁻¹ (ϕ_a = 2.0) at p = 4 MPa, T = 900 K and ω = 2000 s⁻¹. In each fluid particle, hot flame occurs with rapid temperature rise when mf approaches m_{fa}. For every case of mean fuel-mass fraction, the hot flame ignition delay varies over a wide range, because the mixture contains a variety of fuel-mass fractions. However, the particle-to-particle variation of the hot flame start time is not determined by the variation in fuel-mass fraction.

In order to examine the relation between non-homogeneity and temperature rise rate quantitatively, some characteristic times of temperature rise process are defined as shown in Fig. 7. Firstly, the time at 50% temperature rising between the initial and adiabatic temperatures is expressed as τ_{50} and that at 95% as τ_{95}. Next, using these values, temperature rise period τ_i and ignition delay time τ_s are defined as below;

$$\tau_i = 2 \times (\tau_{50} - \tau_{95}) \tag{4}$$
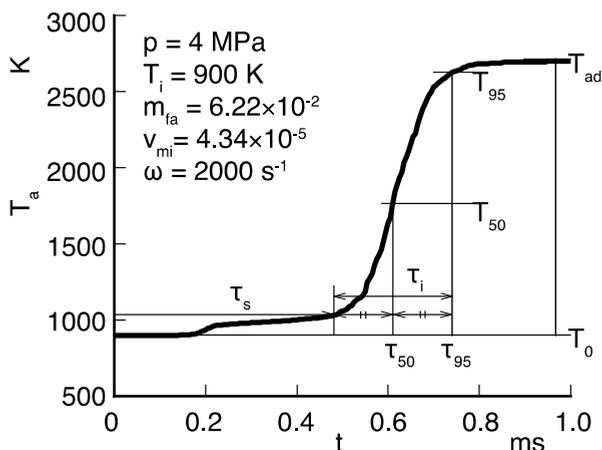
$$\tau_s = \tau_{95} - \tau_i \tag{5}$$

Fig. 7. Definition of ignition delay time τ_s and combustion duration τ_i

Based on these values, the ignition processes of non-homogeneous and homogeneous mixture are discussed. τ_s, τ_{95} and τ_i (marked by an arrow) are displayed for non-homogeneous mixtures with average equivalence ratios ϕ_a of 0.6, 1.0, and 2.0. In Fig. 8, PDF for mf at t = τ_{50} is also displayed for each mixture. In addition, the curves of τ_s and τ_{95} calculated for homogeneous mixtures are drawn. Within this m_f-condition, ignition delay τ_s of the homogeneous mixture becomes shorter with increasing m_f. At the same time, τ_i becomes larger for the leaner and richer sides. Here, ignition delay time of each fluid particle varies due to the variation of equivalence ratio at ignition, therefore, the temperature rise period increases. In addition, m_f dispersion becomes larger for the richer condition, due to shorter ignition delay time. The change in ignition delay against mf

around ϕ_a = 2.0 is small so that τ_i also becomes smaller in spite of the wide distribution of m_f. On the other hand, for the leaner side around ϕ_a = 0.6, the distribution width is narrow, whereas τ_s-change against mf becomes larger than in the stoichiometric case. Then τ_i becomes longer.
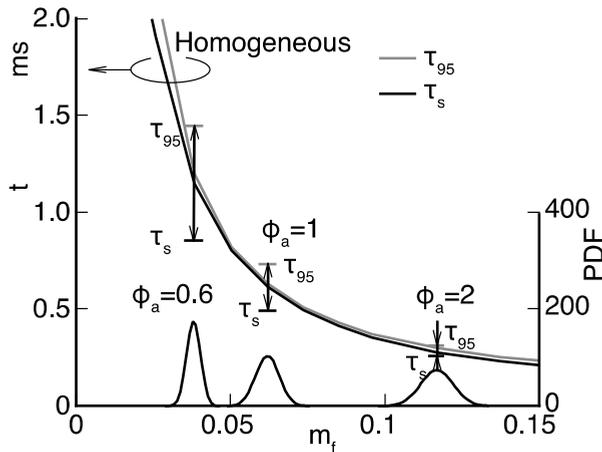

Fig. 8. Changes of ignition delay time τ_s and combustion duration τ_i

In order to confirm the effect of mixing rate on the combustion process, temperature histories are compared for different mixing rates at the same m_f-variance in the middle of hot flame temperature rise, as shown in Fig. 9. The calculation starts with ω = 2000 s^{-1}, then ω is suddenly increased to 10000 s^{-1} just before the ignition. The obtained temperature history differs from that of ω = 10000 s^{-1} and is similar to that of ω = 4000 s^{-1}, whose variance of m_f-PDF at τ_{50} is nearly equal to this ω jumping-up case. This result shows that the rate of temperature rise is strongly affected by the m_f-distribution at ignition rather than the mixing rate. Fig. 10 shows the correlation plots of (a) τ_i–ω and (b) τ_i–v_{m50}. Here, v_{m50} represents the variance of m_f-PDF at τ_{50}. In this case these plots are calculated for wide ranges of v_{mi}, T_i and ω with fixed p = 4.0 MPa and m_{fa} = 6.22×10^{-2} (ϕ_a = 1.0). For the plots on (a), τ_i scatters widely even at the same ω, however, for (b), the plots distribute on a certain curve. This indicates the great influence of m_f variance on combustion duration.

A similar calculation is performed for the case of longer ignition delay and lower temperature rise rate, which is set by m_{fa} = 1.95×10^{-2} (ϕ_a = 0.3), p = 2.0MPa and T_i = 900K.

In this case, the mixture is comparably lean and ambient pressure is low so that τ_s and τ_i become much longer than in the case shown in Fig. 3. Fig. 11 shows the results for ω = 0 s^{-1}, 200 s^{-1}, 400 s^{-1}, 1000 s^{-1} and 2000 s^{-1}. Here, the ignition delay time is longer, therefore, mixture at ignition becomes more homogeneous for the case of ω > 2000 s^{-1}. The ignition delay time τ_s and temperature rise rate become larger with increasing mixing rate, which is similar to the results shown in Fig. 3. Also, τ_{50} is almost constant against ω.
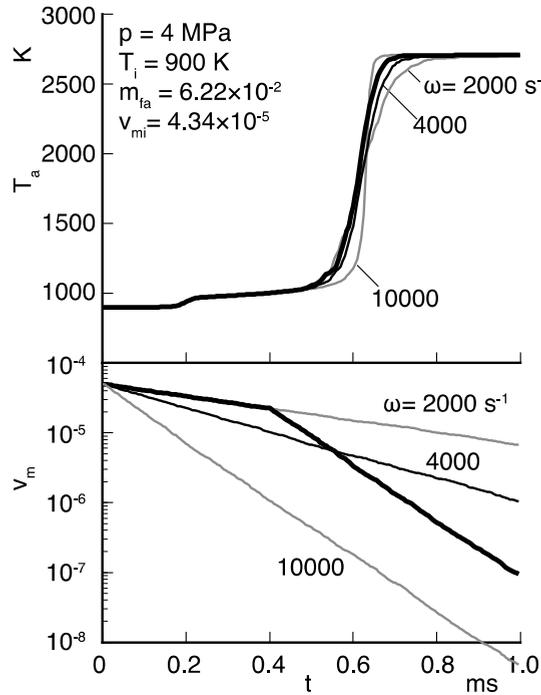
Fig. 9. Effect of collision frequency ω on combustion duration

(a) τ_i-ω Correlation
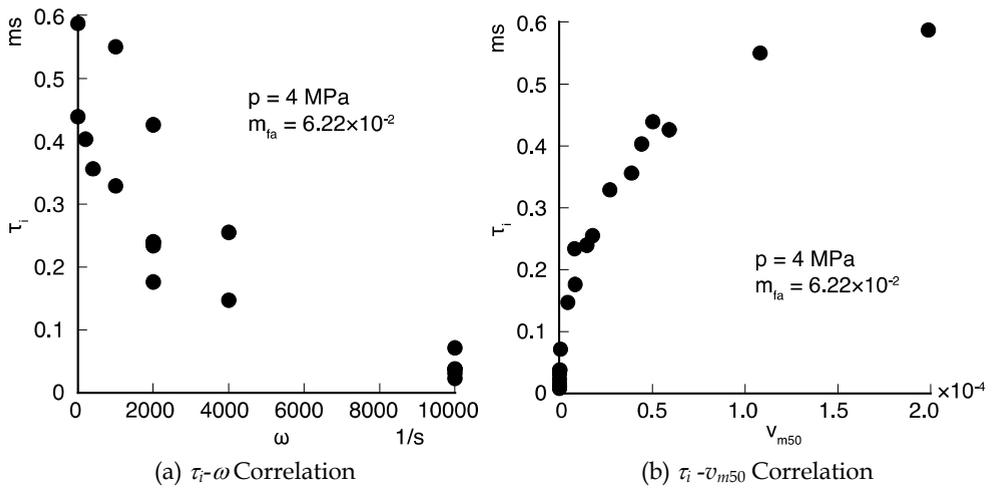
(b) τ_i -v_{m50} Correlation

Fig. 10. Correlations between combustion duration τ_i - collision rate ω and τ_i - mass variation at $t = \tau_{50}$
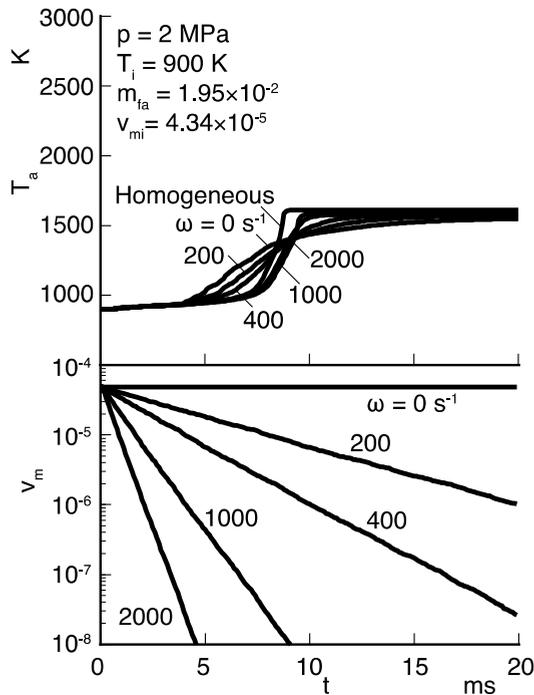
Fig. 11. Courses of mean-temperature T_a and v_m for PCCI-like condition

4. Conclusion

An auto-ignition process of a non-homogeneous mixture in fuel concentration was fundamentally investigated by means of a numerical calculation based on chemical kinetics and the stochastic approach. The auto-ignition process of n-heptane is calculated by means of a semi-detailed mechanism and the non-uniform state of turbulent mixing is statistically described by means of probability density functions and the stochastic method. The following conclusions are derived from the results:

1. For the auto-ignition process of a non-homogeneous mixture during the mixing process, ignition delay time of the cool flame is almost constant against the mixing rate. On the other hand, ignition delay time of hot flame becomes longer with increasing mixing rate. This is because the hot flame ignition delay is more sensible to equivalence ratio than the cool flame delay.
2. For the temperature rising process of hot flame, start points of heat release vary depending on equivalence ratio in a non-homogeneous mixture. Therefore, the rise period increases with increasing non-homogeneity. Also, the temperature rise rate due to heat release of hot flame increases with increasing mixing rate.
3. The tendencies described above are the same for the case of changing equivalence ratio, initial temperature and pressure.

4. Ignition delay time of each fluid particle varies due to the variation of equivalence ratio at ignition, therefore, the temperature rise period increases. In addition, the temperature rise rate becomes larger with decreasing variance of fuel-mass fraction distribution.

5. Nomenclature

C:	Coefficient of modified Curl's model
C_m:	Coefficient of turbulent mixing
f:	Statistical state function
m_f:	Fuel-mass fraction
m_{fa}:	Mean value of m_f
N:	Total number of fluid particles
t:	Time
t^*:	Dimensionless time
p:	Pressure
T:	Temperature
T_a:	Mean temperature
T_i:	Initial temperature
v_m:	Variance of m_f
v_{m50}:	vm at τ_{50}
v_{mi}:	Initial value of v_m
δ:	Delta function
ϕ:	Scalar
ϕ_a:	Mean equivalence ratio
τ_{50}:	Time at 50% temperature rise
τ_{95}:	Time at 95% temperature rise
τ_i:	Combustion duration
τ_s:	Ignition delay of hot flame
ω:	Collision frequency

6. References

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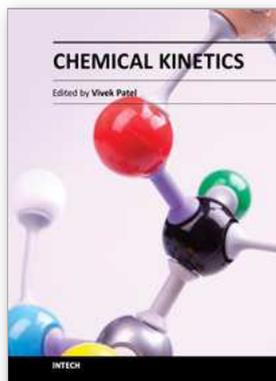
Chemical Kinetics

Edited by Dr Vivek Patel

Chemical Kinetics relates to the rates of chemical reactions and factors such as concentration and temperature, which affects the rates of chemical reactions. Such studies are important in providing essential evidence as to the mechanisms of chemical processes. The book is designed to help the reader, particularly students and researchers of physical science, understand the chemical kinetics mechanics and chemical reactions. The selection of topics addressed and the examples, tables and graphs used to illustrate them are governed, to a large extent, by the fact that this book is aimed primarily at physical science (mainly chemistry) technologists. Undoubtedly, this book contains "must read" materials for students, engineers, and researchers working in the chemistry and chemical kinetics area. This book provides valuable insight into the mechanisms and chemical reactions. It is written in concise, self-explanatory and informative manner by a world class scientists in the field.

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