Numerical Studies on Minimum Ignition Energies of Lean Primary Reference Fuels at Elevated Pressure

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1 Introduction

Ignition and early flame propagation studies does not only contribute to the development of efficient and clean combustion strategies, but are also crucial for safety considerations. Understanding the ignition process and its underlying physical and chemical mechanisms therefore is relevant for various practical applications, including internal combustion engines and example for safety. Minimum ignition energy (MIE), which is the minimum required energy for a successful flame initiation, is an important characteristic for induced ignition [1]. The dependence of MIE on temperature, pressure and mixture composition were studied numerically and experimentally in the past [2–4].

Many applications (e.g. internal combustion engines) operate at elevated pressures, where low-temperature chemistry (LTC) for hydrocarbon fuels like primary reference fuels (PRF) plays a critical role, exhibiting negative temperature coefficient (NTC) behavior closely linked to engine knock [10–12]. Simulations and HCCI experiments have explored LTC's influence on auto-ignition, showing phenomena like cool flames and two-stage ignition, which extend flammability limits [8]. However, the role of LTC for induced ignition of PRFs at elevated pressures is less understood. In particular, its significance for flame regimes, pressure effects on MIE, and RON dependency. To address these gaps, we conduct numerical simulations of flame initiation and propagation in PRF/air mixtures at pressures of 3–25 bar and 373 K, using detailed chemical kinetics and transport processes, aiming to clarify the impact of LTC and its interaction with transport phenomena on ignition.

2 Numerical Methodology

In this study, the ignition and flame propagation process of an initially homogeneous, quiescent mixture of PRF/air is modeled with the in-house code INSFLA [2]. This code solves the conservation equations for a reacting flow in one-dimensional configurations, using detailed chemical kinetics and detailed transport processes. INSFLA uses an adaptive spatial grid and an adaptive time-stepping with error control. A space- and time-dependent ignition source term can be implemented. The simulations use a spherically symmetric domain, which is consistent with our previous study at 1 bar [4]. Symmetric boundary conditions at the center and non catalytic, adiabatic outer boundary conditions for species and temperature are applied. The initial condition for species and temperature are homogeneous fuel/air

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mixtures with an initial temperature $T_0 = 373\,\mathrm{K}$ throughout the entire domain. The investigated pressure range spans $p = 3 - 25\,\mathrm{bar}$, aiming at enhancing the understanding of ignition process at elevated pressure, including the influence of low temperature chemistry. A constant pressure assumption is applied in all simulations because the energy deposition is sufficiently long [2]. The air composition used in this study comprises $21\%\,O_2$ and $79\%\,N_2$.

The investigated fuel in this study is PRF. PRFs are commonly used to determine octane ratings, including both research octane number (RON) and motor octane number (MON). In this paper, we focus on RON and its influence on MIE of PRF, and RON merely characterizes the percentage of iso-octane in the fuel. Lean mixtures are studied here because of their attractive emission reduction and high efficiency. We use the mechanism based on a semi-detailed chemical kinetics model for tolune reference fuels (TRF) with 137 species and 633 reactions for the simulation of PRF [5].

During the ignition duration ($\tau_s = 1 \times 10^{-4}\,\mathrm{s}$), an external source term is introduced. This term is characterized by a power density $\dot{q} = q(r,t)$ which follows an exponential form [2]:

$$\begin{array}{lll} \dot{q} = \dot{q}_{\max} e^{-(\frac{r}{r_s})^8} & \text{for} & 0 < \mathrm{t} \leq \tau_\mathrm{s} \\ \dot{q} = 0 & \text{for} & \mathrm{t} > \tau_\mathrm{s} \end{array} \tag{1}$$

r_s represents the ignition radius. The spatio-temporal profile of the ignition energy density can be found in our previous study [4].

The outcome of the simulations are spatio-temporal profiles of pressure, temperature and chemical composition during and after ignition.

3 Results and Discussions

3.1 Four ignition scenarios

Fig. 1 shows four different types of the spatio-temporal evolution of the temperature during and after ignition. The ignition source, which is active from $t = 0$ to $t = 10^{-4}s$, causes a rise in the maximum temperature for all four cases. The temperature profiles during energy deposition are highlighted in blue.

For Type I, low ignition energy causes only a small temperature rise, and so fails to stimulate any significant chemical reaction in the spark volume. The temperature drop by dissipative effects becomes notable. This corresponds to a failure to ignite the mixture in the spark volume.

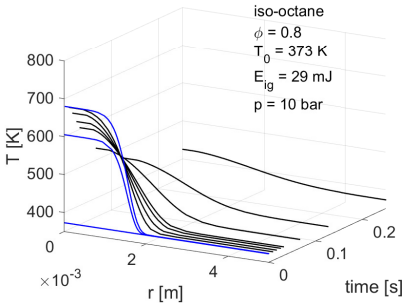
For Type II, a temperature rise about $100\,\mathrm{K}$ is observed after energy deposition. This indicates the formation of a flame kernel inside which reaction commences. The chemical reaction here is only weakly exothermal, and the mixture gets only partially oxidized [6]. This corresponds to the cool flame phenomenon [6]. The flame eventually quenches.

For Type III, a flame kernel with a cool flame emerges like in type II; however, it is followed by another, fast, rise of temperature by about $1500\,\mathrm{K}$ leading to values of completely burned mixture.

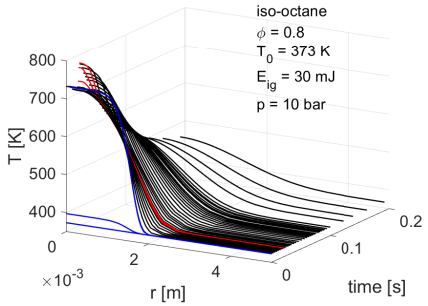
For Type IV, the mixture in the ignition volume is heated up to 1000 K. In this case, no cool flame initiation is observed, the temperature evolves directly to the fully burned state. This type is termed hot flame initiation.

3.2 Dependence of MIE of n-heptane and iso-octane on pressure

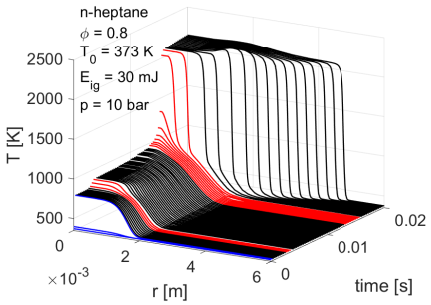
With four different ignition scenarios, the minimum ignition energy (MIE) was determined using a method analogous to interval nesting by identifying the highest non-ignition energy and the lowest



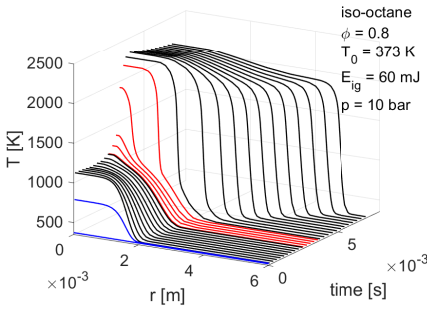
(a) Type I: Ignition failure



(b) Type II: Cool flame ignition and flame extinction

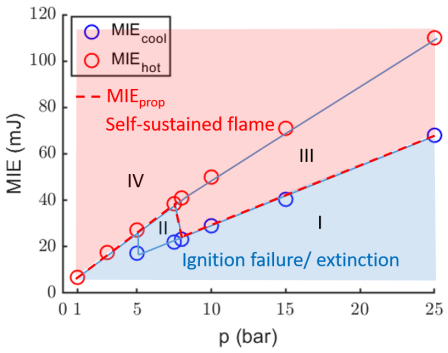


(c) Type III: Two-stage ignition

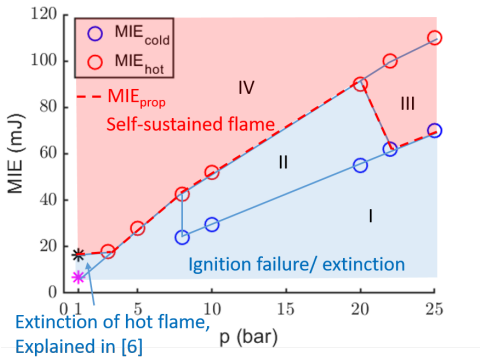


(d) Type IV: Hot flame ignition

Figure 1: Spatio-temperal evolution of the temperature for four different conditions. Red profiles indicate profiles during which ignition occurs



(a) n-heptane



(b) iso-octane

Figure 2: The dependence of MIE on pressure for (a) n-heptane/air and (b) iso-octane/air mixtures, $\phi = 0.8$, $T_0 = 373$K. Red: successful ignition; Blue: extinction

ignition energy to approximate the boundary. $\mathrm{MIE_{cool}}$ and $\mathrm{MIE_{hot}}$ are defined for successful flame kernel formation of cool and hot flames, respectively. $\mathrm{MIE_{prop}}$ (red dotted line) is defined for a self-sustained hot flame, corresponding to flame types III and IV.

Fig. 2 shows the dependence of MIE on pressure for both n-heptane/air mixtures and iso-octane/air mixtures. Ignition types III and IV (which both create a self-sustained flame) are highlighted in red. Ignition types I and II (which do not create self-sustained flames) are highlighted in blue.

Similarly to the simulation results of MIE for hydrogen/oxygen mixtures [2], the MIE for n-heptane and iso-octane hot flames increases with pressure. However, in experiments (see, e.g., [9]), a contrary tendency (MIE decreases with increasing pressure) was observed. The reason is that, in numerical simulations, r_s is kept constant, resulting in a constant V_s. Consequently, the MIE increases as ρ increases with increasing pressure. On the other hand, in experiments, the gap between the electrodes is adjusted at each pressure to reach the MIE. At elevated pressures, ignition can be achieved with a much smaller gap compared to 1 bar. The mass of the mixture in the ignition volume shows a decreasing tendency with increasing pressure, and so does the MIE. In simulations, r_s affects the MIE. As is shown in Fig. 3, at $p = 5$ bar, MIE$= 0.85$ mJ is reached at $r_s = 0.3$ mm. If the ignition radius in simulations is adjusted at each pressure to reach the MIE, the MIE shows a decreasing tendency with increasing pressure, as shown in Fig. 4, which is in agreement with the experimental results.

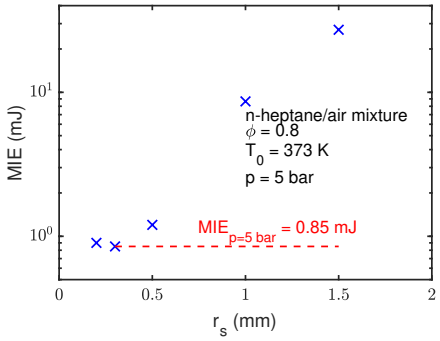


Figure 3: Dependence of MIE on ignition radius for lean n-heptane/air mixtures with $\phi = 0.8$ at $T_0 = 373$ K

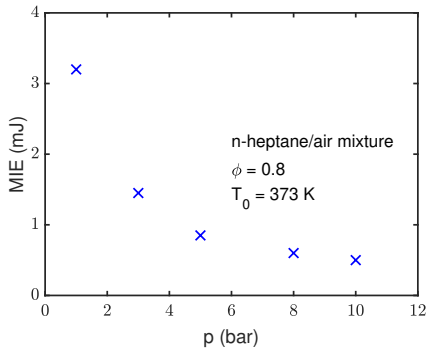


Figure 4: Dependence of MIE on pressure with varying ignition radii for lean n-heptane/air mixtures with $\phi = 0.8$ at $T_0 = 373$ K

$\mathrm{MIE}_{\mathrm{cool}}$ is smaller than $\mathrm{MIE}_{\mathrm{hot}}$ because the temperature required for initiating cool flame reactions is smaller than that for a hot flame. This was also observed for DME/air flames by Ju et al. [8]. Fig. 2 shows that at the same pressure, $\mathrm{MIE}_{\mathrm{hot}}$ and $\mathrm{MIE}_{\mathrm{cool}}$ are comparable for iso-octane/air mixtures and n-heptane/air mixtures. For $\mathrm{MIE}_{\mathrm{hot}}$, this is consistent with the observation in [7], that the MIE of hydrocarbons are similar. For $\mathrm{MIE}_{\mathrm{cool}}$, although the LTC of iso-octane is weaker than LTC of n-heptane, the LTC time scale for iso-octane is still faster than the time scale of heat dissipation, so the $\mathrm{MIE}_{\mathrm{cool}}$ for

iso-octane and n-heptane are comparable. The weaker LTC of iso-octane compared to n-heptane might then lead to cool flame extinction due to heat losses, instead of a two-stage ignition.

At 1 bar, for iso-octane, a hot flame might quench because of strong diffusion. This has been explained in detail in our previous study [4]. At elevated pressure, however, with the same ignition radius as at 1 bar, the extinction of a hot flame is not observed; instead, the hot flame remains self-sustained. This can be understood by noting that at higher pressures, the diffusion is slower and chemical reaction is faster than at lower pressure. Flames propagate at fast speed directly after ignition, and are self-sustaining. At elevated pressure, hot flame extinction might occur with an ignition radius much smaller than at 1 bar.

3.3 Dependence of MIE on RON at elevated pressure

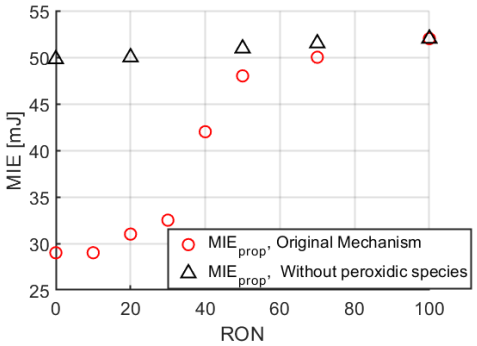


Figure 5: Dependence of MIE on RON at 10 bar, with or without peroxidic radicals, $\phi = 0.8$, $T_0 = 373\,\mathrm{K}$

Fig. 5 shows the dependence of $\mathrm{MIE_{prop}}$ on RON at 10 bar. The $\mathrm{MIE_{prop}}$ at low RONs depends weakly on RON. From RON = 30 to RON = 50, $\mathrm{MIE_{prop}}$ increases rapidly with RON. After that, $\mathrm{MIE_{prop}}$ again depends only weakly on RON. Note that here RON merely characterizes the mixture composition, i.e. stands for the amount of iso-octane in the fuel. With the removal of reactions with peroxidic radicals, $\mathrm{MIE_{prop}}$ becomes independent on RON throughout the whole region. This implies that at 10 bar, LTC which is dominated by peroxidic radicals has a major effect on the dependence of MIE on RON. The dependence of MIE on RON is a result of the slower LTC of iso-octane compared to n-heptane. For n-heptane and PRFs with low RON, the LTC is fast enough for the cool flame to evolve into a self-sustained hot flame. In contrast, for iso-octane and PRFs with high RON, the cool flame quenches, and the ignition energy must reach $\mathrm{MIE_{hot}}$ for a self-sustained flame. In our previous study [4], numerical experiments were conducted for 1 bar. However, the results indicated that removing reactions involving peroxidic radicals did not influence the relationship between MIE and RON. This suggests that increasing pressure enhances LTC, underscoring its significance at elevated pressures.

4 Summary and Conclusion

In this study, induced ignition and early flame propagation in primary reference fuel (PRF)/air mixtures at 3-25 bar are investigated by numerical simulations. The simulations involve a detailed treatment of chemical reactions and molecular transport in a spatially and temporally fully resolved setting, where ignition is triggered by an imposed local energy source. Four qualitatively different types of evolution after the deposition of ignition energy are found: ignition failure, cool flame initiation followed by

flame extinction, two-stage ignition, and hot flame initiation. The results show the significance of low-temperature chemistry (LTC) for the ignition of PRF at elevated pressure. LTC allows a cool flame ignition for temperatures as low as 700-800 K. The cool flame has a lower flame temperature, compared to the hot flame, and only part of the mixture is oxidized. This might cause flame extinction.

$\mathrm{MIE}_{\mathrm{hot}}$ and $\mathrm{MIE}_{\mathrm{cool}}$ are defined for the MIE of a hot flame and a cool flame. They both increase with pressure because of higher density at high pressure. On the other hand, the MIE per mass unit decreases with pressure because diffusive processes get weaker with pressure. Considering the formation of a self-sustained flame, $\mathrm{MIE}_{\mathrm{prop}}$ is defined. For n-heptane, $\mathrm{MIE}_{\mathrm{prop}} = \mathrm{MIE}_{\mathrm{hot}}$ until 8 bar, from there, LTC is strong enough for a cool flame to evolve to a hot flame, $\mathrm{MIE}_{\mathrm{prop}} = \mathrm{MIE}_{\mathrm{cool}}$.

Finally, the dependence of $\mathrm{MIE}_{\mathrm{prop}}$ on RON for elevated pressures is investigated. $\mathrm{MIE}_{\mathrm{prop}}$ is almost independent on RON for small RONs. Above a certain RON, it increases rapidly with RON. After the sudden increase, $\mathrm{MIE}_{\mathrm{prop}}$ is again almost independent on RON.

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