Experimental and Numerical Study of Autoignition/Deflagration Transition Limit in an optical Rapid Compression Machine

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1 Introduction

The development of new efficient and environmentally respectful combustion concepts and systems, capable of using conventional and alternative fuels, has become more important in the recent years. A significant part of the research efforts devoted to reciprocating engines were focused on Low Temperature Combustion (LTC) approaches because of their potential to reduce emissions and achieve higher efficiency compared to conventional combustion methods. In particular, several fundamental studies devoted to compression ignition of homogeneous fuel/air mixtures highlighted the high sensitivity of the global heat release rate to the local combustion mode, such as deflagration, autoignition front and initial detonation, see for instance [1]. In particular, high heat release rate at high load can lead to a rapid autoignition inside the combustion chamber, which causes excessive knocking. This represents a major challenge in HCCI combustion approaches. However only few fundamental studies provide experimental data regarding both the transition between the autoignition front and deflagration, and its sensibility to the thermal gradient in the unburned mixture. This is the objective of the present work, the temperature gradient being both estimated and measured. First, the influence of combustion properties is studied through a numerical approach. To perform a priori estimation of combustion regime, Sankaran [2] criterion is used. Then, the combustion regimes obtained both in the experiment and numerically are analyzed for low values of Ignition Delay (ID< 20 ms). Thus, the experimental results are obtained with similar temperature field topologies, and in particular with well-controlled temperature gradients in the hot core region before the start of the main heat release.

2 Experimental setup and method

Rapid compression machine and protocol: Experiments are performed within an optical Rapid Compression Machine (RCM) developed at Pprime institute [3]. The experiments were carried out for stoichiometric n-heptane/ $O_2/N_2/CO_2$ mixture. Temperature at the top dead center (TDC) is varied by changing the diluent gas composition at fixed volumetric compression ratio ($\varepsilon = 8.9$). A flat piston with a square cross-section with rounded corners (50x50 mm) is used. The duration of compression stroke is

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38 ms approximately. The design of the combustion chamber allows full optical access to the entire volume at TDC through Sapphire or Quartz windows. A detailed description of the apparatus can be found in [3], [4]. In this study, the combustion chamber, cylinder, intake pipes, valves and pressure sensors are heated using a multi-zone regulated heating system in order to obtain a homogeneous or a stratified initial temperature distribution within the cylinder. This leads to low or high temperature gradients after compression in the hottest core region (Compressed Temperature Gradient - CTG). Initial temperature profile along the cylinder axis is measured using a bare bead K thermocouple. Gaseous mixtures are prepared directly inside the cylinder following the partial pressure method. Firstly, the heated combustion chamber and feeding lines are placed under vacuum. N-heptane with high purity grade (>99%) is then injected using microliter syringes and a septum. Pressure of fuel is measured using 0-1300 mbar MKS Baratron 631 capacitive sensor heated at 470 K. Finally, synthetic air (high purity 99.99% O_2, N_2, CO_2) are then admitted through a heated tube to reach the required initial pressure. The operator waits one hour before starting the compression. This is the ideal time slot to guaranty homogenization of the mixture. Pressure level is checked before the test to ensure that the pressure variation is lower than 2%. Pressure evolution is recorded at 100 kHz using a Kistler 6125CU20 piezoelectric sensor coupled to Kistler 5018 amplifier.

Broadband chemiluminescence emissions are recorded using a Photron FASTCAM SA-5 camera, positioned perpendicularly to the cylinder axis. This study focuses on the autoignition behavior at the start of autoignition kernel under known CTG in the hot core region. Due to the thermal stratification eventually generated inside the hot core region, which is not usual in RCM autoignition studies, a specific criterion is selected to determine the ID using chemiluminescence imaging. It corresponds to the start of the combustion kernel formation in the hot core region close to the cylinder head, e.g. at y = 3 mm see Fig. 1b. This instant is close to the very onset of the pressure rise, see Fig. 1 (a). The y = 3 mm location corresponds to the highest temperature before the onset of heat release. In practice, several adiabatic core temperatures can be calculated by considering the different values of the initial temperature profile. Therefore, a homothetic 1D compression is assumed to determine the initial temperature of the fluid which is located at y = 3 mm at TDC. The latter is used as an input to calculate the adiabatic core temperature at y = 3mm at TDC, which is chosen to plot IDs versus $1000/T_{ad}$.

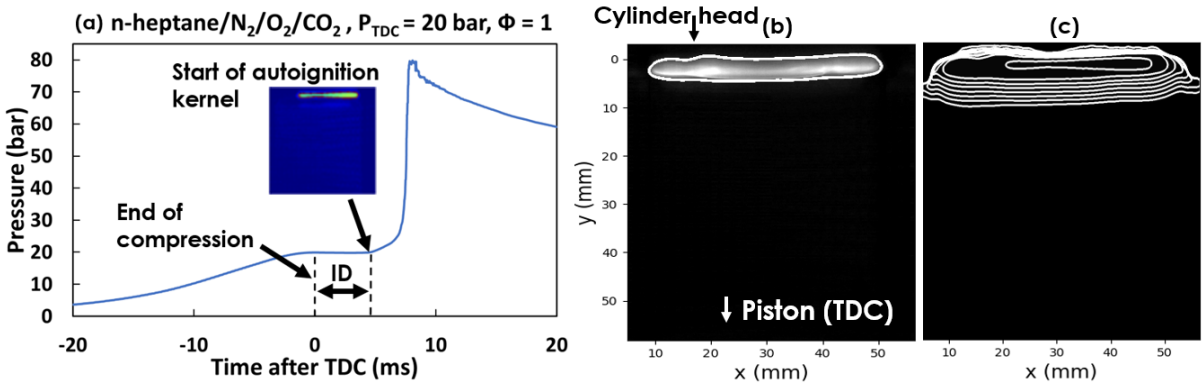

Figure 1: Pressure trace (a), chemiluminescence image and the kernel contour (b), and time evolution of the contours with a time step of 0.23 ms (c). n-heptane - $O_2/N_2/CO_2$ mixtures (21%/59.25%/19.75% by mole in synthetic air) at $\Phi = 1$ and $P_{TDC} = 20$ bar.

The reactive front propagation velocity is measured using chemiluminescence visualization. Chemiluminescence images are first post-processed to detect emission contours. The threshold level is obtained for each filtered image using an anisotropic filter from its intensity histogram (Fig.1 (b)). For more details regarding image post-processing, see [5]. Reactive fronts propagate downwards and their velocity is calculated from the average distance between two consecutive contours divided by the appropriate time step (see Figure 1 (c)). At these early stages of combustion, burned gases expand nearly freely, as pressure variation is kept below 2% in the investigated time range. Velocity values are then

multiplied by the factor ρ_u/ρ_b where ρ_u and ρ_b are the densities in unburned and burned mixture to obtain S_{exp}. Each condition was measured at least four times and the repeatability was found to be very good.

Local temperature gradient measurements: Local temperature gradient was measured using two thin wire Type K thermocouples developed at Pprime institute, located at 3 and 20 mm from the cylinder head, with wire diameters of about 7.6 µm [6]. The junction of these two thermocouples were located at the corner of the dead volume near the cylinder head. This corresponds to the hot core region at the early stages of the post-compression period. The measurements are then compared with theoretical values obtained from same hypotheses as above: the CTG present in the hot core region is estimated from the initial temperature profile (in homogeneous or stratified case) along the symmetry axis of the cylinder, and supposing a homothetic 1D adiabatic compression in the hot core region.

The thermocouples response time depends on the heat convection with the fluid, heat radiation and heat conduction along the wires. The latter depends on the length and diameter of the wire and of the thermal inertia of the prong. Therefore, the measurements were corrected according to Pitts et al. [7] taking into account the temporal velocity profile measured through Particle Image Velocimetry technique at the locations of the thermocouples. After TDC, the measured and corrected temperatures are very close to each other. Therefore, only the raw temperature measurements are considered in the following.

3 Results and discussion

Thermal Gradient measurements: Local CTG are measured using two thin wire thermocouples in non-reactive conditions, e.g. by replacing the 21% of O_2 by N_2 in the synthetic air composition. The results are representative of reactive experiments at the onset of heat release. Thermocouples are localized near a corner of the cubic dead volume, close to the cylinder head (y = 3 and 20 mm). They are vertically aligned, 17 mm away from each other, see Fig. 2. Two different levels of stratification of the initial temperature profile are imposed in the RCM, leading to a low CTG value (1.3 K/mm) measured at the end of compression for the most homogeneous temperature profile, and in a higher CTG (3.7 K/mm) value measured for the most stratified case. The latter is obtained by heating the combustion chamber and the cylinder differently using three zone regulated heating system. Fig. 2 shows the results of temperature measurements for non-reactive mixture during and after compression in this higher CTG case. It shows the presence of a plateau in temperature after the top dead center TDC which corresponds to the homogeneous hot temperature region and confirms that the hypothesis is valid at least 20 ms after TDC in these conditions (see Fig. 2). The CTG based on raw measurements is about 3.7 K/mm at TDC (green line), which is in good agreement with that obtained from adiabatic core temperature values (green dashed line).

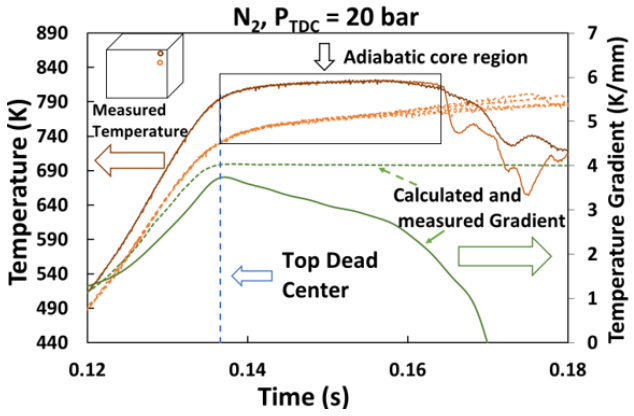

Figure 2: Temperature measurements during and after compression for higher CTG case. Comparison with calculated gradient (green dashed line).

For a given initial temperature profile, the CTG value depends on: 1) the initial temperature profile - two heating conditions are considered is this study -, 2) the temperature at TDC which depends on initial mixture composition - the highest the temperature after compression, the highest the CTG - and 3) on the position in the chamber, as the gradient slightly increases close to the cylinder head. Considering the good agreement above between the measured and the calculated gradient, a slightly higher CTG value is used in the numerical study: it is calculated numerically by reducing the distance between two positions in the adiabatic core region down to 5 mm instead of 17 mm (y = 3 and

8 mm), which corresponds to the free expansion region of the burnt gases. Thus, 4.5 and 2.5 K/mm are retained as CTG value in the hot core region using the two heating conditions and mixture.

Ignition delay measurements: The autoignition of n-heptane/O_2/N_2/CO_2 mixtures is investigated for stoichiometric conditions at 21% of O_2 in synthetic air, 20 bar at TDC. The impact of compressed charge temperature variation on total ID is shown in Fig. 3(a) on Arrhenius type plots as function of the maximum adiabatic core temperature at TDC.

Prior to the analysis of combustion regimes, ID measured with both high and low CTGs are compared. It can be seen from Fig. 3 (a) that the initial temperature profile does not significantly affect IDs values, ID being controlled by the adiabatic core temperature in the hottest unburned mixture region, which is taken into account whatever the initial gradient. The measured IDs are compared to simulations performed using San Diego mechanism [8] and to experimental data issued from the literature.

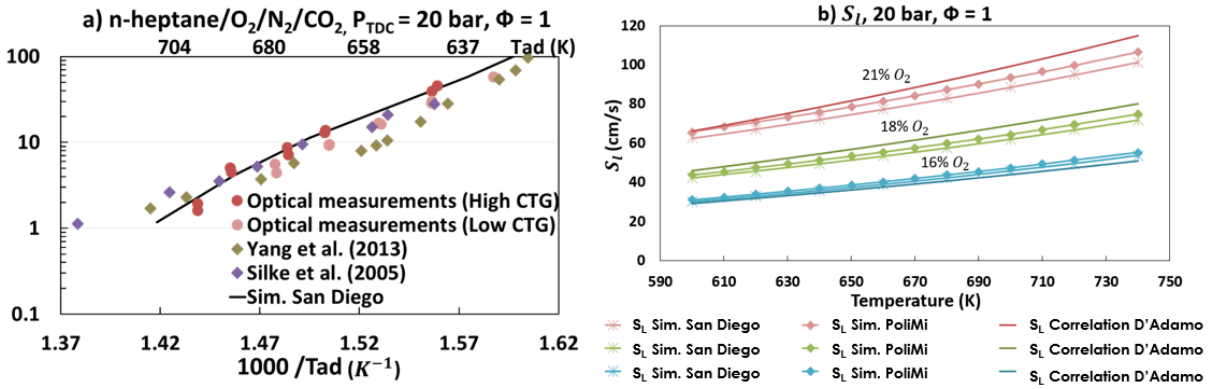

Figure 3: (a) ID measurements as a function of maximum adiabatic core temperature at TDC y = 3mm for n-heptane/O_2/N_2/CO_2 mixtures: measurements at low and higher CTG and comparison to San Diego mechanism and literature at 21% O_2 in synthetic air. b) Laminar flame speed for n-heptane computed using San Diego [8] and PoliMi kinetic schemes [9], comparison with the correlation of D'adamo [10].

Fig. 3 (a) shows that IDs measured in this study are in good agreement with those measured using the RCM of Silke et al. [11] and Yang et al. [12]. In addition, our ID measurements are consistent with those computed using the kinetic scheme of San Diego [8] in homogeneous conditions, taking into account the compression history of our RCM. This shows the relevance of (i) the ID measurements made with a thermally stratified mixture, and (ii) the criteria chosen to estimate the delay and the compressed temperature in these positive temperature coefficient conditions.

Numerical parametric study of autoignition/deflagration transition: A parametric study is performed using numerical tools in order to delineate the regions corresponding to autoignition and to deflagration: different mixture compositions, temperatures and thermal gradient values are considered to this purpose. In the first part, ID computations are performed in adiabatic conditions using a 0D simulation tool based on Cantera. San Diego [8] kinetic scheme is chosen for n-heptane and is validated experimentally in our RCM (see above). Laminar flame velocities S_L were calculated using cantera and San Diego mechanisms for n-heptane. The results – not reported here - show a good agreement with published measurements at low pressure and temperature. The mechanisms were also validated over a large range of equivalence ratios at elevated freestream temperature, but such data are available only at the ambient pressure. Calculation of S_L are made at both high temperature (620-690 K) and high pressure (20 bar) conditions in the following. Nevertheless, the previous validation of the kinetic schemes provides a relatively good confidence in the results. The consistency was also checked by comparing the results with different S_L correlations developed for the high temperature-pressure condition and detailed kinetic scheme for various ranges of temperature and dilution, see Fig 3 (b). After this validation step, the objective is to estimate which combustion regime will occur in the hot core region of the RCM. Simulations for ignition delay times are thus performed taking into account the compression history of the RCM using a non-reactive pressure signal through Cantera. The calculated ID times are then used

to calculate the spontaneous ignition velocity thanks to $S_{sp} = (\frac{d\tau}{dT_0} \cdot \frac{dT_0}{dx})^{-1}$ where τ is the ID time at T_0 [1]. Following Sankaran criterion [2], the S_L to S_{sp} ratio is reported in Figure 4 as a function of molar fraction of oxygen in synthetic air and temperature in the case of low (a) and high (b) compressed temperature gradient for stoichiometric n-heptane/O_2/N_2/CO_2 mixtures at 20 bar at TDC.

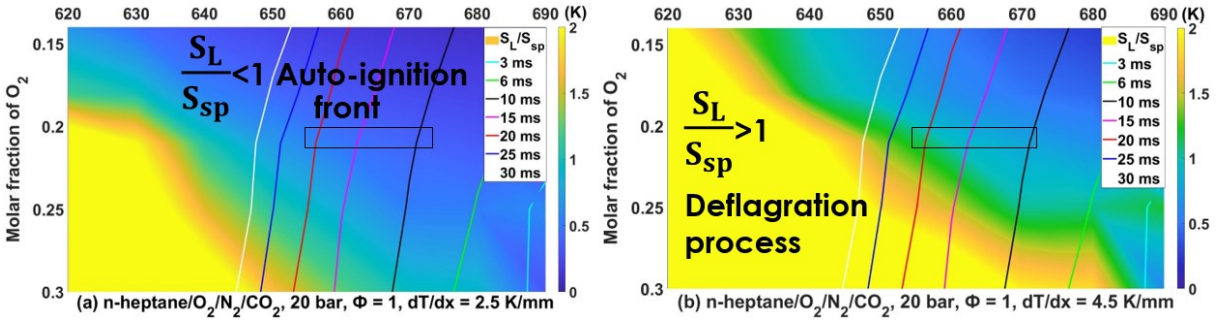

Figure 4: S_L to S_{SP} ratio as function of molar fraction of O_2 in synthetic air and temperature for n-heptane/O_2/N_2/CO_2 mixtures. Φ = 1, P_{TDC} = 20 bar. Low (a) and high (b) CTG (resp. 2.5 and 4.5 K/mm).

Fig. 4 shows that a diluted mixture promotes autoignition. The results are analyzed for identical values of ID (iso-lines in the graphs): in the low CTG case of 2.5 K/mm, high ID times (>20 ms) are required to transit from autoignition to deflagration, by changing molar fraction of oxygen. Nevertheless, only short IDs (<20 ms) will be considered for the experiments, so that the hot core region remains sufficiently large during the autoignition process in our flat piston RCM. On the other hand, in the case of high CTG of 4.5 K/mm (Fig. 4 b), deflagration region is wider. This condition is pertaining to study the transition between combustion regimes in our RCM, since for each ID value chosen between 6 and 20 ms, both deflagration and autoignition are possible depending on the oxygen molar fraction. Influence of equivalence ratio and pressure is also studied; the results are not shown here for the sake of brevity.

Experimental confirmation of the numerically predicted combustion regimes: The reactive front velocity S_{exp} is measured from chemiluminescence records. In this respect, the investigated time range is chosen so that pressure variation is lower than 2% during the reactive front propagation. Ignition delays are kept lower than 20 ms to ensure a sufficiently wide hot core region (Δy~20 mm). In Figure 5, S_{exp} values obtained from chemiluminescence records at 21% O_2 are compared to (i) the theoretical value of the autoignition velocity S_{sp} and (ii) to the unstretched laminar flame speed S_L. The same method as described above is used to obtain numerically S_L and S_{sp}. In our study, temperatures ranging from 650 to 700 K at TDC (y = 3 mm) are investigated experimentally. This corresponds to ignition delays lower than 20 ms, see Fig. 3a. Two cases are reported: high and low CTG, respectively plotted in red and purple. For each of them, S_{sp} is plotted for both the highest and lowest estimate of CTG (resp. solid line, dashed lines) for each initial temperature profile and adiabatic core temperature at TDC. CTG is calculated using 1D compression hypothesis evoked above and the temperatures at y = 3 and 8 mm.

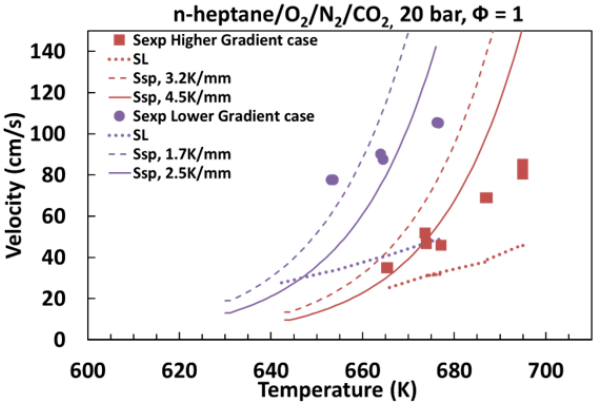

Figure 5: Comparison between S_{exp}, S_{sp} and S_L in the case of low CTG (Purple color) and high CTG (Red color) as function of adiabatic core temperature at TDC. 21% of O_2 in synthetic air.

The case of high CTG (red plots in Fig. 5) is first analyzed: at temperature lower than 680 K (ID higher than ~10 ms), S_{exp} values obtained from chemiluminescence record are close to both the S_L and the

calculated autoignition front velocity S_{sp}. This confirms the numerical finding of Fig. 4, where S_L to S_{sp} ratio are found to be between 0.8 and 1.2 (see rectangle in Fig 4 b), e.g. the conditions are close to the limit between deflagration and autoignition. Respectively, at higher temperature > 680 K (ID < 10 ms), S_{exp} is greater than S_L, but close to S_{sp}, which confirms the numerical finding, e.g an autoignition regime is found in these conditions. In the case of lower CTG values (purple color in Figure 5), S_{exp} values are also relatively close to S_{sp}, and significantly higher than the laminar flame speed S_L. This is consistent with the numerical findings: only autoignition fronts propagate in these lower CTG conditions (see rectangle in Fig 4 a).

Conclusion: The characteristics of combustion regime transitions are investigated in a fully optical RCM. An original combination of experimental conditions is set to generate quasi-1D autoignition fronts or deflagrations propagating inside a wide laminar and hot region. At this location, temperature gradients are estimated and also measured with thin thermocouples of 7.6 µm. The transition limits between combustion regimes are predicted numerically using Sankaran criterion and calculation of S_{sp} and S_L by changing thermal gradients and mixture composition. The predicted combustion regime is consistent with the velocity of the reactive front measured from chemiluminescence emissions. In particular, conditions are found for which an elevated Compressed Temperature Gradient in the core region (CTG) leads to a transition to deflagration while a low CTG promotes autoignition. This confirms the relevance of the proposed approach for fundamental studies of combustion regime transitions.

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