An Experimental Investigation of Iso-octane Ignition Phenomena

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Abstract

High-speed digital imaging has been used in rapid compression facility (RCF) studies to investigate ignition phenomena of iso-octane/air mixtures. Sequential images were captured for each experiment. The results indicate the existence of two ignition regimes. In one domain, ignition is rapid, typically less than 76 μsec, and ignition occurs simultaneously throughout the test volume. In the other domain, reaction fronts form and propagate within the test volume prior to volumetric ignition. The data span equivalence ratios from $\phi = 0.20$ to 1.98, with inert/O$_2$ gas ratios from 1.38 to 5.89, pressures from 8.7-16.6 atm and temperatures from 903-1020 K. The transition between the two regimes is discussed in the context of the mixture composition and experimental conditions. The analysis shows that the fuel mole fraction is a key parameter dictating the boundary between the modes of ignition. Below a critical mole fraction limit, volumetric ignition is observed; above the critical limit, reaction fronts are consistently present prior to volumetric ignition. The ignition delay times for both ignition regimes are well reproduced using a homogenous simulation with detailed reaction chemistry, when the state conditions are modified to account for the presence of the reaction fronts. The results are also discussed in terms of theory proposed for modes of ignition.

Keywords: ignition, ignition delay time, iso-octane, reaction front, rapid compression facility
1. Introduction

Intermediate-temperature, premixed, lean fuel conditions are of considerable importance in many advanced combustion strategies which focus on simultaneously reducing pollutant emissions and increasing combustion efficiencies, such as homogeneous charge compression ignition (HCCI) engines [1] and integrated gasification combined cycle (IGCC) power plants [2]. The reaction kinetics often play a significant role on the combustion characteristics that control performance of the devices based on lean premixed technologies. As a consequence, there have been significant efforts to investigate the chemical kinetics of fuel/air mixtures at conditions relevant to these combustion applications (e.g. [3,4,5,6,7,8,9]). The uniform high-temperature, high-pressure conditions created by shock tubes and rapid compression facilities (RCF) make these apparatuses ideal for isolating the reaction chemistry and quantifying the ignition behavior of the fuel/air mixtures over a broad parametric space, including equivalence ratio and dilution. Ignition data are also critical as a means to validate and refine detailed, skeletal, and reduced chemical reaction mechanisms.

During the course of several fundamental experimental [10,11,12,13,14,15] and computational [16,17] ignition studies of hydrocarbon and hydrogen fuels, spatially-resolved phenomena have been observed that indicate the existence of multiple ignition regimes in addition to volumetric ignition. For example, during recent RCF ignition studies of iso-octane mixtures [3,4], the presence of reaction fronts prior to volumetric ignition was observed at certain conditions, while volumetric ignition with no spatially-resolved features occurred at other conditions. Gu et al proposed 5 regimes of ignition or propagation modes in computational ignition studies when hot-spots were introduced into otherwise homogeneous mixtures of hydrogen and hydrogen and carbon monoxide with air [16]. Gu et al. proposed that the regimes
are delineated by the local temperature gradient, the critical temperature gradient, and the acoustic velocity of the test gas mixture. Similarly, Sankaran et al. identified two ignition regimes (spontaneous explosion and flame-like deflagration) in their computational study of ignition of hydrogen/air mixtures in turbulent scalar fields of temperature gradients [17].

Studies of engines operating in HCCI mode have also indicated the presence of reaction fronts at conditions below the lean flammability limit, where flames are not considered self-sustaining [18,19,20,21]. The presence of reaction fronts at lean conditions can have critical effects on ignition timing and pollutant emissions [18,19] and on safety considerations if traditional flammability limits, such as the trends developed by Zabetakis [22], are applied.

The objective of the current work was to identify and quantify the characteristics and modes of ignition of lean, intermediate-temperature iso-octane/air mixtures and to provide a quantitative understanding of the chemical and physical mechanisms that are important in the ignition regimes. The technical approach used was to measure the ignition delay times for iso-octane mixtures over a broad experimental domain using a rapid compression facility and simultaneously use high-speed digital imaging to capture the ignition phenomena. The results are discussed in terms of proposed reaction chemistry and ignition theory.

2. Experimental Approach

The rapid compression facility

All experiments were conducted in the well-controlled and well-characterized environment of the University of Michigan Rapid Compression Facility (UM RCF). The ignition behavior of iso-octane/air mixtures was investigated in terms of several parameters including: equivalence ratio, oxygen concentration, fuel concentration, temperature, and pressure. High-speed digital
imaging, and pressure and emission time-histories were used to characterize the ignition experiments. A detailed description of the UM RCF, the operating procedure, and the results of benchmark experimental studies characterizing performance can be found in Donovan et al. [23], He et al. [3,4] and Donovan [24].

Briefly, the UM RCF consists of five major components: the driver section, the driven section, the test manifold, the sabot (free piston), and the hydraulic control valve assembly. The driver section (154 mm i.d.) contains pressurized air and is separated from the driven section (2.74 m long, 101.2 mm i.d.) via the fast-acting hydraulic globe valve assembly and a scored sheet of plastic (0.05 mm, Mylar®). The driven section is a long stainless steel tube with a honed and chromed interior surface which is connected to the test manifold. The sabot has a replaceable nosecone which is made of ultra-high molecular weight polyethylene. The body of the sabot is solid (Delrin®) and contains a copper counterweight to balance the nosecone. U-ring seals (virgin Teflon® with stainless steel radial springs) eliminate gas blow-by past the sabot.

A schematic of the test manifold and imaging system is shown in Fig. 1. The four main components of the test manifold are the convergent section, the extension section, the instrumented test section, and a transparent end wall. The stainless steel convergent section bridges the 101.2 mm bore of the driven section to the 50.8 mm bore of the remainder of the test manifold components. The total length of the extension section is variable by design (through combinations of subcomponents) to yield different compression ratios. The test section has a length of 50.6 mm, and is equipped with two optical ports for laser access, a pressure transducer port, and two additional instrumentation ports. For the current study, the test section was instrumented with a piezoelectric transducer (Kistler 6041AX4) and charge amplifier (Kistler 5010B) for pressure measurements. The end wall seals the test manifold and allows optical
access to the test volume.

Two end walls, quartz or polycarbonate, were used to provide optical access to the test manifold. The quartz end wall (with a diameter of 78 mm and a thickness of 45.25 mm) has a double layer anti-reflective coating on both faces. The coatings are optimized for transmission of visible wavelengths and the window has excellent reflection- and glare-reduction properties. The polycarbonate (Lexan®) end walls are polished (140 × 140 mm square with a thickness of 12.5 mm) and have no coatings. Peak transmission for the polycarbonate end walls occurs at visible wavelengths. When the polycarbonate windows were used, the windows were replaced regularly (every 5-10 experiments) in order to maintain high image quality.

The test gas mixtures were made in a dedicated mixing tank, and the mixture composition was determined by measurement of the relative partial pressures of the gas-phase reactants. The specific heats of the mixtures were controlled through the balance of argon, carbon dioxide, and nitrogen as inert gas diluents. The specific heat of the mixture was adjusted and the experimental compression ratios were varied to achieve the targeted temperatures and pressures.

Prior to each experiment, the sabot is placed at the upstream end of the driven section, downstream of the hydraulic valve assembly and the scored plastic sheet. The RCF is evacuated. The driven section and test manifold are then charged with the test gas mixture, and the driver section is charged with high-pressure air. The hydraulic valve is opened, breaking the plastic sheet, and launching the sabot down the length of the driven section. The test gas mixture is compressed ahead of the sabot. The sabot comes to rest in the test manifold when the nosecone seals (via an annular interference fit) with the extension section. The core region of the test gases is sealed in the test manifold while the cooler gases near the walls are sealed in the shoulder region of the convergent section.
High-speed imaging details

The large size of the quartz and polycarbonate end walls allows the entire test volume to be imaged using a high-speed color digital video camera (Vision Research, Phantom V7.1, 800 × 600 pixel SR-CMOS 48 bit color array, capable of 150 kHz at reduced spatial resolution). A fast 50 mm lens (f/0.95 Navitar TV Lens) and c-mount extension tube were used with the camera to optimize the capture of available light emission.

Each time the camera settings were changed (e.g. different experimental conditions, frame rate or spatial resolution), the camera sensor array was recalibrated. The camera was first aligned with the test manifold approximately 40 cm from end wall, and the camera was focused on a plane inside the test section, 2 cm from the end wall. To maximize the capture of the emission from the test section and limit the depth of field, the wide aperture lens was used in the fully open position. The camera frame rate and spatial resolution were selected at this time. The camera sensor was then calibrated using a black reference (where the camera lens was covered) and the zero signal level of each pixel in the CMOS array was assigned.

For this study, the high-speed digital camera was used to acquire continuous full-frame video sequences of the ignition experiments at speeds from 10,000 to 60,000 frames per second (fps). The frame rate has an inverse relation to the total spatial resolution, and at 26,000 fps (the typical frame rate used), the spatial resolution was maintained at the maximum allowable setting of 256 × 256 pixels. These settings resulted in each frame corresponding to 38 µsec, and each pixel in the CMOS array imaging focused light from a volume with a height × width × depth of approximately 198 µm × 198 µm × 2 mm.

The camera was triggered by a circuit designed to output a pulse to the trigger input of the
camera. The signal to the triggering circuit was supplied by an unamplified photo-detector (Hamamatsu S1787-12) located on the driven section of the RCF. Emission from a laser diode (TIM-201-3, 3 mW, 650 nm) was directed onto the active element of the photo-detector such that the laser emission was orthogonal to the path of the sabot. When the sabot passed the laser diode, the change in signal from the photo-detector triggered the camera. Using this arrangement, images were acquired throughout the compression and ignition processes.

3. Results

Typical pressure and pressure derivative data for RCF iso-octane ignition experiments are shown in Figs. 2 and 3. Results for conditions where volumetric ignition occurred and where reaction fronts preceded volumetric ignition are presented in each figure, respectively. In both data sets, the initial pressure rise is due to compression of the test gas mixture ahead of the sabot. At the end of compression, the pressure reaches the first maximum. This time is set as \( t = 0 \) sec and is labeled \( P_{\text{max}} \) in the figures. The pressure then decreases slightly due to cooling losses to the test volume walls. After a delay period, the mixture auto-ignites resulting in a rapid increase in pressure.

Figure 4 presents a selection of the frames from the imaging sequence corresponding to the volumetric ignition data of Fig. 2. As seen in Fig. 4, blue emission occurred uniformly throughout the test volume with increasing intensity until maximum emission levels were observed at \( t = 10.260 \) ms. No structures or spatial non-uniformities were observed in the emission images for this condition, and ignition occurred simultaneously throughout the test volume. The peak in the emission intensity occurred at the same time as the peak in the pressure derivative.
Frames from the imaging sequence corresponding to the pressure data of Fig. 3 are shown in Fig. 5. The images show the presence of discrete regions of reaction in the test volume. The reaction fronts are observed early in the image sequence \( t = 6.098 \) ms, and they propagate throughout the test volume until volumetric ignition is recorded at \( t = 22.5 \) ms. The effect of the propagation of the discrete zones of reaction is apparent in the pressure time history. The pressure data show a slow increase at times consistent with the formation and propagation of the reaction fronts, prior to the rapid increase in pressure associated with volumetric ignition (see Fig. 3). The expansion of the reaction fronts serves to further compress the unignited test gas mixture and consequently the pressure and temperature in the test section gradually increase prior to volumetric ignition. On the contrary, the pressure data corresponding to volumetric ignition (Fig. 2) show little pressure rise during the delay period prior to the ignition.

In previous UM RCF studies of iso-octane ignition [3,4], the effective pressure \( (P_{\text{eff}}) \) for each experiment was defined as the time-integrated average pressure from the maximum pressure \( (P_{\text{max}}) \) due to compression to the minimum pressure \( (P_{\text{min}}) \) due to cooling after compression, but before ignition, i.e.

\[
P_{\text{eff, min}} = \frac{1}{(t_{P_{\text{min}}} - t_{P_{\text{max}}})} \int_{t_{P_{\text{min}}}}^{t_{P_{\text{max}}}} P \cdot dt
\]

If this definition is applied to the conditions where reaction fronts were present, the pressure at the time of volumetric ignition is underestimated. In order to account for the effects of the reaction fronts, the effective pressure was defined for each experiment in this work as the time-integrated pressure between the point of maximum pressure due to compression \( (P_{\text{max}}) \) and the point of maximum rate of pressure rise \( (dP/dt_{\text{max}}) \), or

\[
P_{\text{eff, dP/dt}} = \frac{1}{(t_{dP/dt_{\text{max}}} - t_{P_{\text{max}}})} \int_{t_{P_{\text{max}}}}^{t_{dP/dt_{\text{max}}}} P \cdot dt
\]
Equation (2) has little effect versus Eq. (1) on \( P_{\text{eff}} \) determined for the conditions where only volumetric ignition was observed. For example, for the data presented in Fig. 2, \( P_{\text{eff},p_{\text{min}}} \) is equal to \( P_{\text{eff},dP/dt} \). More generally, the average difference between \( P_{\text{eff}} \) defined using Eqs. (1) and (2) was less than 0.5% for experiments where no reaction fronts were observed. The average difference in \( P_{\text{eff}} \) using Eqs. (1) and (2) for conditions where reaction fronts were present was less than 7%.

The effective temperature for each experiment was determined as in previous UM RCF studies [3] using the effective pressure and by numerical integration of the isentropic relation:

\[
\int_{T_i}^{T_f} \frac{\gamma}{\gamma-1} \frac{\gamma}{\gamma-1} d\ln T = \ln \left( \frac{P_{\text{eff}}}{P_o} \right)
\]

where \( P_o \) is the charge pressure, \( T_o \) is the initial temperature (typically 298 K), and \( \gamma \) is the temperature-dependent ratio of the specific heats of the unreacted test gas mixture, which is determined using the NASA thermodynamic data base [25]. The average difference in \( T_{\text{eff}} \) due to the definitions of Eqs. (1) and (2) for \( P_{\text{eff}} \) is 0.1% when reaction fronts were not present and 1.5% when reaction fronts were present.

For each experiment, the ignition delay time (\( \tau_{\text{ign}} \)) was determined using the pressure time-history. In previous UM RCF studies of iso-octane mixtures [3,4], the ignition delay time was defined as follows. The end of compression was designated as the first time the pressure reached \( P_{\text{eff}} \) due to compression, using \( P_{\text{eff}} \) defined by Eq. (1). The portion of the pressure trace which corresponded to the sharp pressure rise due to ignition was linearly extrapolated. The relatively constant portion of the pressure trace prior to ignition was also linearly extrapolated. The intersection of the two linear fits was designated the start of ignition. The ignition delay time was defined as the difference between the end of compression and the start of ignition.
When reaction fronts are present, this definition for $\tau_{\text{ign}}$ is not feasible. The gradual pressure rise during the ignition delay period complicates linear extrapolation of the pressure data. Additionally, $P_{\text{eff}}$ defined using Eq. (2) leads to pressures higher than $P_{\text{max}}$ for some conditions. Consequently, in this work the ignition delay time was defined as the time between $P_{\text{max}}$ and $dP/dt_{\text{max}}$. The definition for $\tau_{\text{ign}}$ is illustrated in Figs. 2 and 3. When compared with the previous data for $\tau_{\text{ign}}$, the new definition for $\tau_{\text{ign}}$ yields an average difference of $\pm 2\%$.

For each experiment where reaction fronts were observed, the rate of propagation of the reaction fronts, $U \ [\text{m/s}]$, was determined from the image sequences, as shown schematically in Fig. 6. Note that in order to make the presence and propagation of the reaction fronts clearer, Fig. 6 presents data from the same imaging file as that of Fig. 5 where a threshold filter has been applied to create binary images. The propagation rates were determined by tracking features on the reaction surface through time. The result is an estimate for the propagation rate in the direction normal to the surface of the reaction front. The image analysis indicated that the highest propagation rates of the reaction fronts were associated with propagation into the unconstrained environment towards the center of the test volume (as opposed to propagation into gases bounded by the test chamber walls). Only the maximum velocities are reported here.

The general features were the same for each of the categories of ignition. For conditions where reaction fronts were observed, more than one reaction front was typically present. Reaction fronts did not consistently initiate in the same location of the test volume, i.e. in some cases the fronts started in the center or top of the test volume as compared to the data of Fig. 5. For conditions where only volumetric ignition was observed, ignition (as indicated by homogeneous blue emission) was rapid and typically occurred in less than 76 µsec.

A summary of the iso-octane ignition data is presented in Table 1. The table shows the range
of conditions and mixture compositions examined, with equivalence ratios varying from $\phi = 0.2$ to 1.98, inert gas/O$_2$ ratios varying from 1.38 to 5.89, pressures varying from $P_{\text{eff}} = 8.7$-16.6 atm (determined using Eq. (2)), and temperatures varying from $T_{\text{eff}} = 903$-1020 K. The imaging and pressure data for each experiment were designated as one of the two categories of ignition described above: volumetric ignition or reaction front propagation/volumetric ignition. The volumetric ignition data are identified by a “V” designation in the table.

The ignition delay time determined for each experiment is also provided in Table 1, along with the maximum propagation rate for each experiment where reaction fronts were present. Calculated reaction front velocities ranged from 0.5 to 13.7 m/s, much faster than laminar flame speeds for iso-octane at elevated temperatures and pressures, and far below the speed of sound (a, based on the unreacted gas composition at $T_{\text{eff}}$ and $P_{\text{eff}}$) for the mixtures, which are provided in Table 1.

In order to identify trends in the $\tau_{\text{ign}}$ data, regression analysis is required to isolate the effects of each parameter of interest, e.g. temperature. In a previous UM RCF study of (only) volumetric ignition of iso-octane mixtures [3], regression analysis yielded the following best-fit ($R^2$ value of 0.98) correlation for the ignition delay time data:

$$\tau_{\text{ign}} = 1.3 \times 10^{-4} P^{-1.05} \phi^{-0.77} \chi_{O_2}^{-1.41} \exp(33700/R_{[\text{cal/mol/K}]}T)$$

where $P$ is pressure [atm], $T$ is temperature [K], $\phi$ is the equivalence ratio, $\chi_{O_2}$ is the oxygen mole percent, and $\tau_{\text{ign}}$ is the ignition delay time [ms]. Using Eq. (4) to normalize the data as necessary, a summary of the ignition delay time measurements from the current study is presented in Fig. 7 as a function of inverse temperature.

Figure 7 provides a comparison of the data using the definitions for effective pressure based on Eqs. (1) and (2). As noted earlier and demonstrated in Fig. 7, the data where only volumetric
ignition was observed are relatively unaffected by the definition for $P_{\text{eff}}$. However, when Eq. (1) is used to define $P_{\text{eff}}$ for the reaction front data, the data are highly scattered and show poor agreement with the volumetric ignition correlation. When the effects of the reaction fronts on $P_{\text{eff}}$ are considered, using the larger integrated time domain of Eq. (2), there is excellent agreement between the data from the two ignition regimes, as seen in Fig. 7.

Due to the revised definitions for $\tau_{\text{ign}}$ and $P_{\text{eff}}$, differences in the range of conditions studied in the work by He et al. [3] and the current work, and the two modes of ignition considered here, the need for a new correlation for $\tau_{\text{ign}}$ was investigated. Regression analysis was conducted on the complete $\tau_{\text{ign}}$ data set of Table 1, using exclusively $P_{\text{eff}}$ based on Eq. (2). Multiple forms to the expression were considered yielding similar qualities of fit. For consistency, the same form as Eq. (4) is presented here, with a best-fit $R^2$ value of 0.92.

$$\tau_{\text{ign}} = 2.8 \times 10^{-3} P^{-1.25} \phi^{-0.79} \chi_{O_2}^{-1.14} \exp(27300/R_{[\text{cal/mol/K}]}T)$$

Although the fit parameters differ from those of Eq. (4), the new correlation yields only slight changes in the predicted values for $\tau_{\text{ign}}$, as seen in Figs. 8-11.

Summaries of the effects of temperature, pressure, equivalence ratio, and oxygen mole fraction on $\tau_{\text{ign}}$ are presented in Figs. 8-11, respectively, using $P_{\text{eff}}$ defined based on Eq. (2), and where the data have been normalized as necessary using Eq. (5). Equation (5) is provided for reference in each of the figures as the solid line. The volumetric and reaction front $\tau_{\text{ign}}$ data are in excellent agreement with each other for each of the parameters considered, and there are no systematic deviations in the trends indicated by the two ignition regimes. The recommended uncertainty in the ignition delay time measurements (presented as the error bars in Fig. 8) is $\pm 13\%$ and is based on the standard deviation of $\tau_{\text{ign}}$ relative to the correlation provided in Eq. (5).
Trends in the rate of propagation of the reaction fronts as a function of the state conditions and the mixture properties were also investigated. The analysis indicated no strong correlation with temperature, pressure, equivalence ratio, or oxygen mole fraction, and even extremely fuel lean mixtures were capable of sustaining a reaction front. However, there was a strong dependence of \( U \) on fuel mole fraction (\( \chi_{\text{fuel}} \)) as seen in Fig. 12, where \( U \) increases nearly linearly with increasing \( \chi_{\text{fuel}} \). When compared with conditions where only volumetric ignition was observed, the data of Fig. 12 imply the existence of a critical fuel mole fraction limit, \( \chi_{\text{fuel,crit}} \), above which reaction fronts are self-sustaining, and below which only volumetric ignition is observed. Using a linear fit to the \( U \) data and extrapolating to \( U = 0 \) m/s, yields a value for \( \chi_{\text{fuel,crit}} \) of 0.4.

4. Discussion

The experimental data for \( \tau_{\text{ign}} \) were compared with predictions using a single-zone model and the detailed chemical reaction mechanism developed for iso-octane by Curran et al. [26]. For the modeling, the AURORA™/CHEMKIN™ 4.0 suite of programs [27] was used assuming a homogeneous, adiabatic, constant volume system. Using the initial conditions of the unreacted mixture composition, \( P_{\text{eff}}, \) and \( T_{\text{eff}}, \) the corresponding predictions for ignition delay time are provided in Table 1 as \( \tau_{\text{pred}}. \) The trends based on the mechanism by Curran et al. are also provided for comparison in Figs. 8-11. As seen in Table 1 and Figs. 8-11, the predictions for \( \tau_{\text{ign}} \) are in excellent agreement with the experimental data (within an average of 11% for the volumetric ignition data, and within an average of 15% for the reaction front data). The ability to accurately reproduce \( \tau_{\text{ign}} \) for the conditions where reaction fronts were observed using a homogeneous model is an important finding, which indicates the primary effects of the reaction fronts on the unignited reactants is compression. Transport effects, which are localized and are
not captured by the single-zone model used, do not appear to significantly influence $\tau_{\text{ign}}$.

Ignition theory supports the existence of regimes where reaction fronts can be sustained prior to volumetric ignition. The classification of ignition regimes between thermal explosion and detonations in the presence of non-uniform initial conditions was first proposed by Zeldovich [28]. He describes the regimes based on the relative values of the laminar flame speed ($u_l$), the rate of spontaneous propagation of the reaction fronts ($U$ in the current work, $u_{\text{sp}}$ in [28]), the speed of sound, and the rate of normal detonation ($u_d$). The results of the current work are in the 3rd category described by Zeldovich, where $u_l < U \ll a < u_d$. Zeldovich stated that in this regime, the propagation of reaction fronts is slow such that “the pressure has sufficient time to equalize,” and the reaction fronts do not depend on the “heat conductivity” of the mixture, which is consistent with the observations made here.

It is important to note the formation of the reaction fronts are not due to cool flame or negative-temperature coefficient (NTC) chemistry. Iso-octane does not exhibit NTC behavior at the pressures and temperatures considered here [4]. Additionally, when NTC behavior is observed, the effect on the ignition delay time for iso-octane is weak. Lastly, model studies using the reaction mechanism by Curran et al. [26] confirm that accelerated ignition due to low temperature chemistry does not occur at the range of conditions encountered during the RCF experiments conducted in this study.

The initiation of the reaction fronts is likely due to small thermal non-uniformities in the test section, as proposed by Zeldovich [28]. Characterization studies of the UM RCF indicate the variability in the temperature field in the core region is typically less than 10 K [23]. The extent of the thermal inhomogeneities is primarily determined by the motion of the fluid off the nose-cone of the sabot and the mixing of the cooler fluid near the wall. Consequently, the temperature
gradients that exist in each experiment are expected to be of the same magnitude when the sabot velocity profiles are similar, as they were in this study. However, the reaction fronts are only self-sustaining at specific compositions. Thus, the thermal gradients may be necessary for initiation, but they are not sufficient to sustain the propagation of the reaction fronts.

Gu et al. [16] have shown in studies of reaction front propagation in H2 and CO mixtures that very small temperature gradients, e.g. ΔT = 0.17 – 1.7 K, from hot spots ranging from 1 to 3 mm in diameter are sufficient to have marked impact on the mode of reaction front propagation. The authors defined the dimensionless temperature gradient, ξ:

\[ \xi = a \left( \frac{\partial T_o}{\partial r} \right) \left( \frac{\partial \tau_{ign}}{\partial T_o} \right) \]  

(6)
as a means to quantify the boundaries between ignition regimes. In Eq. (6), T_o is the temperature of the surroundings of the hot spot and r is radius of the hot spot. Values for ξ for the current work were estimated using the data of Table 1, the temperature derivative for τ_{ign} based on Eq. (5) and assuming a 10 K gradient associated with a 3 mm hot spot. The values for ξ did not indicate a clear limit between the ignition regimes. However, the dimensionless ratio of the speed of sound to the measured maximum rate of propagation (a/U) supports the proposition of a limit based on fuel mole fraction. Figure 13 presents a/U as a function of χ_{fuel} for the experiments where reaction fronts were observed. At low values of χ_{fuel}, the ratio exponentially approaches a limit consistent with the value χ_{fuel,crit} = 0.4 that was estimated based on the data of Fig. 12.

It is interesting to compare the critical limit for fuel mole fraction determined in this work with the correlations for flammability limits developed by Zabetakis [22]. Zabatakis surveyed a large body of experimental flammability data and compiled the results as a function of fuel type. For paraffin hydrocarbon fuels such as iso-octane, he proposed that the lean flammability limit
(LFL, volume percent basis) is not a function of pressure, and the LFL for propagation into high-
temperature premixed fuel/air mixtures can be determined by extrapolating LFL measurements
made at room temperature. Specifically, Zabetakis’s recommendation for \( C_nH_{2n+2} \) fuels in air
was
\[
\chi_{LFL(T'\,C)} = \chi_{LFL(25'\,C)} - \frac{0.75}{\Delta H_c} (T - 25'\,C)
\]
where \( \chi_{LFL} \) is the fuel mole fraction on a percent basis at the lean flammability limit and \( \Delta H_c \) is
the net heat of combustion [kcal/mol]. Using an average temperature of 682 °C (955 K) for the
current data set, \( \Delta H_c = 1305.3 \) kcal/mol for iso-octane at 25 °C [29] and \( \chi_{LFL(25{\,}C)} = 0.95 \) (the
recommendation for \( n \)-octane [22]), Eq. (7) yields a value of \( \chi_{LFL(682{\,}C)} = 0.57 \), which is in
good agreement with the estimate of \( \chi_{fuel,crit} = 0.4 \) based on the U data of Fig. 12.

It is interesting to note that Kaiser et al. [18] observed a dramatic change in the engine-out
emissions at \( \chi_{fuel} \cong 0.3 \) in their study of a single-cylinder engine operating in HCCI mode using
gasoline fuel. Kaiser et al. proposed that the shift in emissions could be explained by flame
“propagation over relatively short distances” for fuel mole fractions less than 0.3. The results of
the current study support that hypothesis.

5. Conclusions

The current work has demonstrated the existence of two distinct ignition regimes for iso-
octane at high pressures and intermediate temperatures. The regimes are consistent with ignition
theory and the results serve as the first experimental data to capture and quantify the
characteristics of the modes of ignition. For the range of conditions studied, the regimes are
delineated by a critical fuel mole fraction. The results for ignition delay time indicate the
primary effect of the reaction fronts is to increase the pressure in the test chamber and thereby accelerate volumetric ignition. A homogeneous model and the reaction mechanism for iso-octane by Curran et al. [26] yields predictions for $\tau_{\text{ign}}$ that are in excellent agreement with the experimental data. As many advanced combustion methods utilize lean, low-temperature premixed strategies at similar conditions to those studied in the current work, the data provide both valuable insight into anticipated ignition performance as well as quantitative data critical to validating and refining ignition theory.

Acknowledgements

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Table 1. Summary of experimental conditions and results. The mixture composition is provided on a mole basis. The equivalence ratio is based on iso-octane to O$_2$ molar ratios. Experiments where only volumetric ignition was observed are denoted V.

<table>
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<th>Φ</th>
<th>Inert / O$_2$</th>
<th>Test gas composition$^a$</th>
<th>P$_{eff}$$^b$ [atm]</th>
<th>T$_{eff}$ [K]</th>
<th>U [m/s]</th>
<th>a$^c$ [m/s]</th>
<th>Ignition delay time $^e$ [ms]</th>
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<td></td>
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Fig. 2. Typical pressure and pressure derivative time-histories for iso-octane ignition experiments where no indications of reaction fronts are observed prior to volumetric ignition. Experimental conditions are $\phi = 0.30$, and inert/O$_2$ = 5.00. The effective temperature based on the effective pressure $P_{\text{eff}} \cdot \frac{dP}{dt} = 9.0$ atm is $T_{\text{eff}} = 1020$ K.

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Fig. 4. Imaging sequence corresponding to the data of Fig. 2 and conditions of volumetric ignition where $\phi = 0.30$, $T_{\text{eff}} = 1020$ K, $P_{\text{eff}} = 9.0$ atm, inert/O$_2$ = 5.00, $\tau_{\text{ign}} = 10.3$ ms, 26,000 fps (no color adjustment). Note not all frames in the imaging sequence are presented. The time interval spanned for these frames is shown in Fig. 2.

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Fig. 6. Example of the image analysis used to determine the reaction front propagation rates. The images are presented in a binary form to enhance clarity. The vector indicates the direction used to determine the speed and is set as orthogonal to the surface of the propagating reaction front. The images are from the data set shown (in part) in Fig. 5, where $\phi = 0.20$, $T_{\text{eff}} = 917$ K,
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**Fig. 9.** Summary of reaction front and homogeneous ignition data for iso-octane ignition delay time as a function of pressure. The experimental data have been normalized to \( T = 980 \ \text{K}, \ \phi = 0.4, \ \text{and} \ \chi_{\text{O}_2} = 21\% \) using Eq. (5). Equation (4) and the results of model predictions for \( \tau_{\text{ign}} \) based on the reaction mechanism of Curran *et al.* [26] are provided for comparison.

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References


