A Rapid Compression Machine Study of n-Decane Ignition at Intermediate Temperatures

V.V. Leschevich, O.G. Penyazkov, S.Yu. Shimchenko
Physical and Chemical Hydrodynamics Laboratory, Heat and Mass Transfer Institute
220072, Minsk, Belarus

1 Introduction

n-Decane is one of the normal alkanes that are often chosen as the component of surrogate jet and diesel fuels [1]. Studies related to the auto-ignition behaviors of various surrogate neat components are of fundamental and practical importance because it is the underlying phenomenon for the combustor design of most engines and ignition delay data have extensive uses in the development and validation of combustion reaction mechanisms. Experiments on the ignition of n-decane have been carried out previously and most of them were conducted in the heated shock tube at high temperatures [2–8]. Probably only in one of these studies ignition delay times of n-decane/air mixtures were measured within a wide range of temperatures 650–1300 K [2]. Data from this paper show a negative temperature coefficient (NTC) in the Arrhenius plot and afterward were used as kinetic targets for most kinetic mechanisms of n-decane oxidation. Although Zhukov et al. [5] reported ignition delay time data at temperatures below 1000 K, his measurements are limited in the range of high pressures. The NTC behavior was found for experimental data at temperatures 790–1000 K and pressures about 8 MPa for stoichiometric mixtures and the depth of pothole is even larger in comparison with data of Pfahl et. al [2] at a pressure of 5 MPa. The remaining data of Zhukov et. al. received at 1–1.3 MPa for stoichiometric mixtures and at 8-10 MPa for lean mixtures follow the linear inverse temperature dependence of logarithms of the ignition delay times at temperatures higher than 1000 K. Using a rapid compression machine (RCM) the ignition of n-decane was studied only by Kumar et al. [9]. The ignition delay times for lean and rich mixtures were obtained in the temperature range of 635–770 K and pressure range of 0.7–3.0 MPa. Thus there is a lack of experimental data for intermediate temperature range, which does not allow to investigate the temperature dependence of the ignition delay times of n-decane. The objective of this study is to extend the range of available experimental data for n-decane auto-ignition, with special emphasis on temperatures ranging from 800 to 1000 K. Experiments were conducted using a rapid compression machine (RCM) for homogeneous stoichiometric n-decane/air mixtures at pressures ranged from 0.6 to 1 MPa in order to sew the new data together with the data obtained by shock tube [7] at high temperatures. The same study was previously carried out with methane-air mixtures [10]. It was showed the unavoidable presence of contaminating ultrafine particles in the test volume, as well as their capability to be auto-ignited and to provoke earlier ignition of gas mixtures. Therefore, the high-speed imaging

Correspondence to: V.Leschevich@dnp.itmo.by

of ignition and combustion processes was performed during each experiment of this study in order to detect the influence of contaminating particles on time and spatial uniformity of n-decane ignition.

2 Experimental procedure

The RCM is a single-shot, piston-cylinder compression device compresses a given mass of premixed gaseous mixture to the desired final pressure and temperature in a short interval of time (20–40 ms). The compressed gas temperature at the end of compression or top dead center, TDC, is achieved by altering the compression ratio by adjusting the stroke length. The pressure at TDC can be independently controlled by varying the initial pressure of the reactants. The RCM applies a flat piston without a creviced shape and details about the RCM design and operational features can be found elsewhere [10]. Since the current experiments are conducted for a relatively low vapor pressure fuel, the combustion chamber walls and its associated feed lines are maintained at an elevated temperature (about 60 C) to prevent condensation of fuel. A homogeneous stoichiometric mixture of n-decane vapor and air (O_2 – 20.73%, N_2 – 77.93%, $C_{10}H_{22}$ – 1.34%) was prepared in a heated stainless steel tank of known volume. The liquid fuel component (about 70 μl) is added under ambient conditions to the vacuumed (<13 Pa) tank on a gravimetric basis using burette. Then volume is filled than with synthetic air (20.9% O_2 and 79.1% N_2) from other vessel to pressure needed for stoichiometric composition. This pre-mixture is subsequently heated up to a temperature of 60 C at which the partial pressure of the fuel vapor in the mixture is well below its saturation value. The heating is greater than 5 h, to enable complete vaporization of the liquid component and mixing with air. The cylindrical 50 mm in diameter reaction chamber is equipped with a Kistler 6031U18 pressure sensor combined with a Kistler 5015A charge amplifier, a valve providing access to the reactor chamber for feeding reactants and a quartz windows enabling side and end optical access. Through side wall window (dia. 5 mm) light emission during ignition and combustion was recorded by two photomultiplier tubes, PMT. Interference filter transmitting the wavelength of 430.8 nm was installed in front of one PMT photocathode for detecting emission of exited CH, while the second PMT with photosensitivity in the wave range of 300-600 nm was used for overall light registration. The quartz window (dia. 50 mm) in the end of the compression cylinder was used for high speed imaging of ignition and combustion processes by «LaVision HighSpeedStar X» camera at frequency of 12,500 frames per second and resolution of 1024 × 1024.

3 Results and discussion

Typical experimental pressure and photomultipliers signals are shown in Fig. 1. Signals of four experiments are merged on one plot by the end of the compression stroke (t=0) that corresponds to the time moment of the maximum pressure rise. All these experiments were conducted under the same initial conditions (13 kPa and 333 K) and adjustments of RCM (compression ratio 21±0.1). The calculation of ideal adiabatic pressure and temperature at the end of compression based on the compression ratio assuming an isentropic process gives values of 0.706 MPa and 860 K. As it is seen from experimental records in Fig. 1 the pressure measured at the TDC varies from test to test and is higher than adiabatic value. The TDC temperatures derived from the measured pressures at the start and end of compression according to “adiabatic core” hypothesis exceed ideal adiabatic value on 30-70 K. The opposite is usually observed as a consequence of heat losses and boundary layer effects. It can be caused by several reasons. First of all, when piston assembly quickly stops from high velocity (up to 15 m/s) compression piston makes several longitudinal vibrations near the end position. It causes a short-time increase of the compression ratio and therefore of the final pressure and temperature. But it can’t fully explain the observed discrepancy because about 4 mm displacement has to occur in order to compensate the entire excess of pressure. From another hand, it is can be attributed to the exothermic reaction occurring in the final stages of compression [11]. Especially it is pronounced for reactive mixtures that exhibit two-stage ignition. As was found for n-decane ignition [9] the first-stage low-pressure rise is relatively insensitive to temperature variations and is able to quickly initiate even under modest conditions. For example, it took about 1 ms for lean (ϕ=0.8) n-decane/air mixture at 14.3 bar and 697 K [9]. This temperature (calculated from pressure traces in Fig. 1) is

achieved at about 3.5 ms before the TDC in experiments 3 and 4. It can explain the more rapid pressure rise as well as shorter ignition delay time in experiment 3 and ignition during compression in experiment 4. Moreover, in experiments 2 and 3, a short-time flash of light was detected by PMT at 2 ms before the end of compression while the light emission from excited CH radicals was detected significantly later. The same flash was detected in experiment 1 but at the TDC. In all experiments, the luminescence of CH radicals is in the agreement with pressure growth caused by combustion even when light from all volume was collected through end window by lenses on the photocathode of PMT (experiment 4). The end and side window light observations are well correlated. It means that this flash appears closer to the end of compression cylinder. No light was registered by a high-speed camera at these time moments perhaps as a result of sensitivity limitation of the CCD sensor. Even when the pressure in test chamber begins to grow after ignition delay time pass low-intensity images of reaction zone are registered (Fig. 2). The time moment of each frame (exposure time is 79 μs) are marked in the Fig. 1 by squares and vertical lines of color as pressure signals. Volumetric ignition was observed simultaneously in several places of the combustion chamber and subsequent ignition process of the rest volume looks like volumetric. In addition, there were no direct observations of burning contaminating particles at this time in the test chamber. They appear later when combustion zone of n-decane extend to the entire volume.

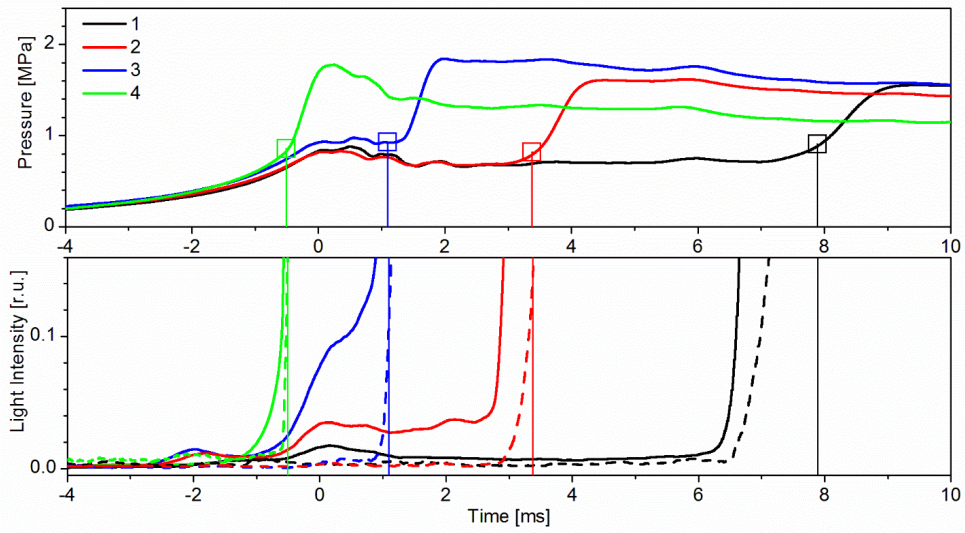

Figure 1 Top: pressure; bottom: light (solid lines) and CH radical (dashed lines) emission registered in five experiments for stoichiometric n-decane/air mixture at 860±1 K

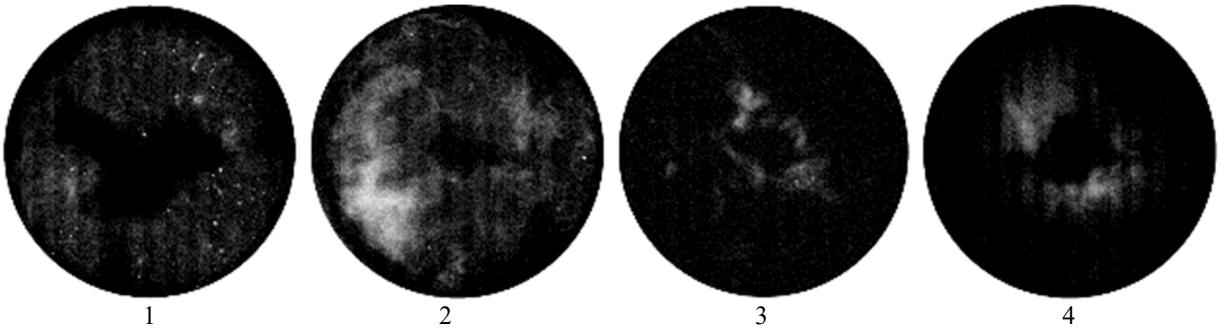

Figure 2 Images registered at the start of n-decane ignition in experiments 1-4 from Fig.1

The same tendency of n-decane ignition was found at lower compression ratio (19.63±0.04) that corresponds to ideal adiabatic pressure of 0.648 MPa and temperature of 845 K when compression starts from 13 kPa and 333 K. The longest ignition delay is observed when the lowest flash appears near the end of compression (experiment 3 in Fig. 3). The combustion of some contaminants (first frame in Fig. 4) in test chamber was found at the time moment corresponded to the high-intensity flash registered by PMT at 1.6 ms after TDC in experiment 2 presented in Fig. 3. Probably, it caused more rapid ignition of n-decane-air mixture although it has been started in other places of the combustion chamber (frames 2-4 in Fig. 4).

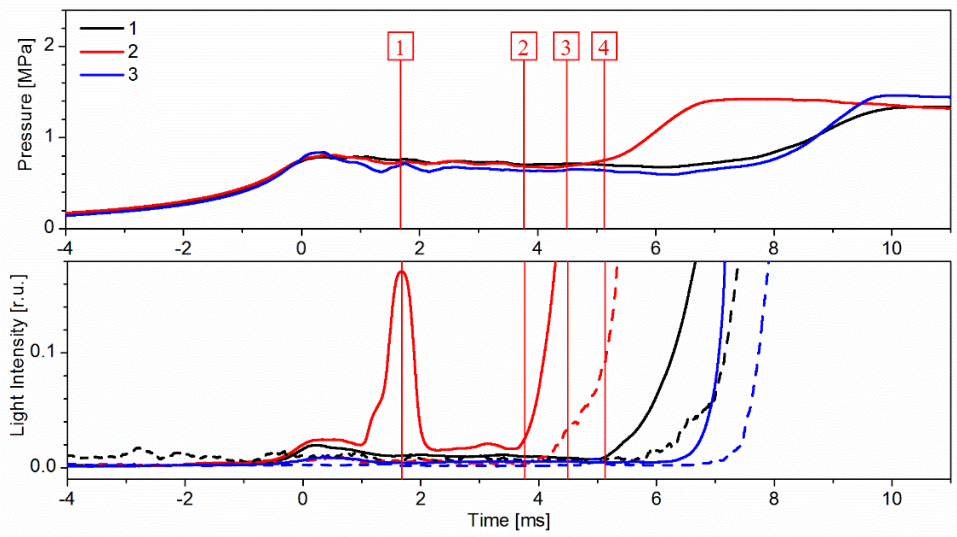

Figure 3 Top: pressure; bottom: light (solid lines) and CH radical (dashed lines) emission registered in three experiments for stoichiometric n-decane/air mixture at 845±1 K

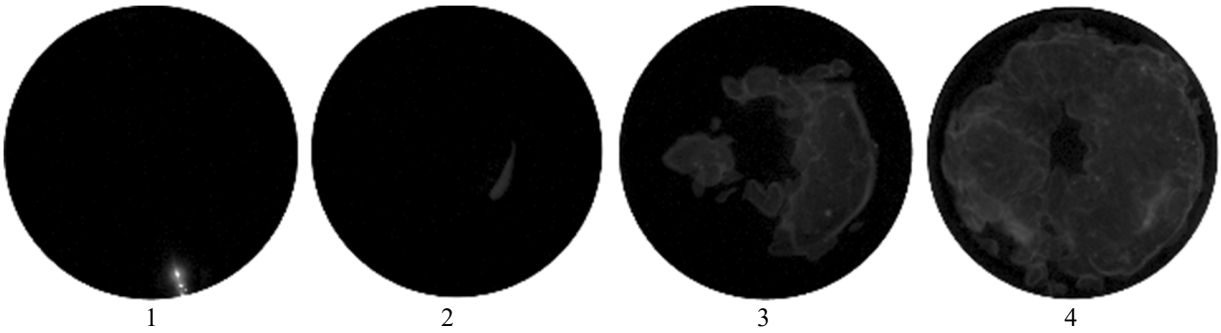

Figure 4 Frame sequence registered at the n-decane ignition in experiment 2 from Fig.3

At compression ratio 18 (adiabatic temperature 825 K) ignition of stoichiometric n-decane/air mixture was not observed. However, PMT registered single flashes of light and high-speed imaging discovered single burning dust particles at these time moments. At compression ratio higher 22 (adiabatic temperature 870 K) ignition of n-decane starts always during compression, therefore, ignition delay times were obtained in a narrow temperature range in this study. Moreover, as it was discussed there is a problem with a definition of temperature at the end of compression. In order to maintain consistency throughout this paper and compare results with other literature data, the ideal adiabatic value is quoted as the reference temperature. The difference between this value and temperature derived from the measured pressures at the start and end of compression is reported as an associated

error bar in the Arrhenius plot for the temperature. Ignition delay time was determined by pressure measurements as the time difference between the peak pressure at the end of the compression stroke and the beginning of the pressure rise caused by the ignition of the mixture (5% of the maximum value). The longest of times measured during multiple runs under identical conditions is selected as representative data (e.g. experiment 1 in Fig. 1) and compared with the reported ignition delay data of stoichiometric n-decane/air mixtures in a somewhat similar pressure range (0.7–1.4 MPa) in Fig. 5. Measurements from [9] have been reported for lean and reach mixtures, making it difficult to directly compare these results to the current work, but they are presented on the plot because there are no other data obtained by RCM facility. Similarly, shock tube data from [5] and [6] at high pressures are shown in Fig. 5 due to lack of results at the temperature range of current study.

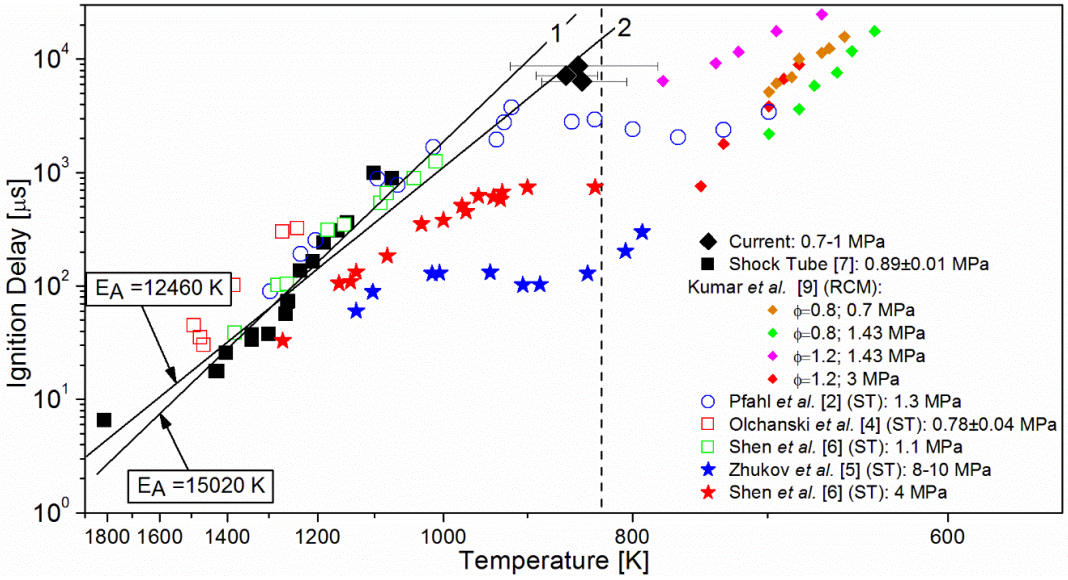

Figure 5 Comparison of measured ignition delay times for stoichiometric n-decane/air mixture with the literature data

The obtained RCM data complement well as an extension of the high-temperature shock tube data [7] for the same mixture composition and similar density. Measurements of [2] at the post-shock pressure of 1.3 MPa and the same temperatures show shorter ignition delay times and this discrepancy has to be even more significant if consider differences in experimental methods and ignition delay time definitions. Probably it can be attributed to higher pressures because measurements at 4 MPa from [6] and at 8-10 MPa from [5] show the tendency of drastically reduction of ignition delay times with increasing pressure. This behavior was also seen in our measurements when a pressure at the end of the compression stroke has been slightly varied. But any relationship has not been established because of the narrow range of pressure changing and discussed the challenge of the temperature interpretation. With regard to RCM data of [9] as an extension of our current RCM and previous shock tube results [7], a better agreement is found for measurements at 1.43 MPa and =0.8 in an assumption of a monotonically increasing trend of ignition delay with a decrease in temperature. Moreover, taking into account the absence of ignition at lower temperature (marked by a vertical dashed line in Fig. 5) and no evidence of the onset or the inclusion of the NTC-like behavior for ignition delay for stoichiometric n-decane/air mixture with decreasing temperature is seen in the current study. The observed deviation from a linear fit of shock tube data (line 1 in Fig. 5) can be attributed to the experimental uncertainty of temperature definition as well as to restriction of RCM application for measurements at intermediate-to-high temperature conditions connected with a possibility of chemical reactions during compression. The general temperature dependency of ignition delay times measured by shock tube and RCM (line 2 in Fig. 5) follows Arrhenius law with a global activation energy of E_A= 12460 K. This value well agrees with the deduced in [9] activation temperature T_A=12198 K for the total ignition delays.

4 Concluding remarks

Ignition of n-decane in the air was investigated in RCM at pressures varied from 0.6 to 1 MPa for stoichiometric mixtures within the temperature range of 820– 900 K. Volumetric and insensitive to burning contaminating particles ignition was observed simultaneously in several places of the combustion chamber. The significant difference of ignition delay times measured in multiple runs under identical conditions was found. The end and side wall light emission observation showed that more rapid ignition can be attributed to low-intensity light that can appear in the combustion chamber at different time moments in repeated experiments. Due to sensitivity limitation of CCD sensor of high-speed camera, this light was not visualized on images and, therefore, its source was not established. The longest of ignition times measured from repeated runs were selected as representative data and compared with the reported literature data. Current data are well jointed with previously obtained at high temperatures shock tube data for the similar mixture density and together exhibit a monotonically increasing trend of ignition delay with a decrease in temperature. No evidence of the onset of the NTC-like behavior for ignition delay for stoichiometric n-decane/air mixture is seen for studied range of pressures.

Acknowledgements

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