Full Length Article

Autoignition and flame spectroscopy of propane mixture in a rapid compression machine

T. Goyal, D. Trivedi, O. Samimi Abianeh*

Mechanical Engineering Department, Wayne State University, 5050 Anthony Wayne Drive, Detroit, MI 48202, United States

ABSTRACT

The autoignition process of propane has been studied using an optically accessible rapid compression machine (RCM). The ignition delay times of the fuel were measured at two compressed pressures of approximately 21 and 30 bar, equivalence ratios of 0.5, 0.8, 1.0 and 1.2, and at various compressed gas temperatures. A detailed kinetic model was also used to simulate the autoignition. A new methodology was also developed and discussed to include the effect of heat transfer in the mechanism model simulation. The combustion process imaging was acquired using high-speed camera and the deflagration like combustion with blue continuum spectrum was observed close to the end of ignition delay at all of the studied conditions, excluding the condition with very long ignition delay (e.g., 90 ms). The deflagration concluded with detonation for most of the studied conditions. The excited radicals' chemiluminescence emissions were acquired using flame spectrometer and Electron-Multiplying Charge-Coupled Device (EMCCD) with wavelength ranging from 280 nm to 700 nm. Several excited radicals were detected during autoignition process and their time-resolved intensity as a function of mixture conditions (pressure, temperature and equivalence ratio) were studied.

1. Introduction

Experimental measurements and modeling of propane are available in the literature due to importance of the propane kinetic model as a sub mechanism for heavier hydrocarbon kinetic model. Considering the ignition delay timings, these studies can be classified in three categories: systems using static reactors which are mostly concerned on low temperature regime, temperature less than 650 K, e.g., [1]; secondly, studies focusing at relatively high temperatures with a short ignition delay times of approximately less than 2 ms using shock tube, e.g., [2]; finally, there are studies focusing on low temperature and negative temperature coefficient (ntc) region with considerably longer ignition delay measurements using flow reactor and rapid compression machine, e.g., [3]. Some of the major propane studies, regarding ignition delay measurements, kinetic development and speciation, using Shock tube, Jet Stirred, Flow Reactors, and rapid compression machine (RCM) are reviewed in this section.

1.1. Propane studies using shock tube

Burcat et al. (1971) [4] measured propane ignition delay times behind the reflected shock waves over a pressure range of 2–10 atm, temperature range of 1250–1600 K and equivalence ratios from 0.125

to 2. Mathematical correlation was also developed showing the influence of oxygen, argon and propane concentrations on ignition delay, which shows argon has no effect on ignition delay times.

Dagaut et al. (1992) [5] developed a new detailed kinetic model of propane oxidation. The detailed mechanism is able to reproduce experimental species concentration profiles from high-pressure jet-stirred reactor ($900 \leqslant T/K \leqslant 1200$; $1 \leqslant P/atm \leqslant 10$; $0.15 \leqslant \phi \leqslant 4$) and in a turbulent flow reactor at 1 atm, ignition delay times measured in shock tube ($1200 \leqslant T(K) \leqslant 1700$; $2 \leqslant P(atm) \leqslant 15$; $0.125 \leqslant \phi \leqslant 2$), H-atoms concentrations measured in shock tube during the pyrolysis of propane and burning velocities of freely propagating premixed propane-air laminar flames.

Lamoureux et al. (2002) [6] measured ignition delays for alkanes (methane, ethane and propane) using shock tube at pressure range of 0.1–1.8 MPa, temperature range of 1200–1700 K and equivalence ratios of 0.5–2.0. For each alkane, a correlation between ignition delay time, temperature, pressure and concentration was developed with an accuracy better than 20% for all of the measurements.

Cadman et al. (2000) [7] utilized shock tube to investigate and measure ignition delays of propane over a wide range of pressures (5–40 bar), relatively low temperature range (> 850 K) and at fuel lean to stoichiometric conditions. The experimental data was used to validate a mechanism from study of Dagaut et al. (1992) [5]. The used

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kinetic model could not fully mimic the measured ignition delay times at temperature below 1000 K.

Brown and Thomas (1999) [8] measured propane ignition delay using shock tube at pressures from 1.3 to 5 atm and temperature range of 1073–2211 K by monitoring CH emissions from ethylene and propane mixtures using eight spark Schlieren camera in the test section window. The influence of argon and nitrogen as diluents on ignition delay was also investigated. They concluded that the effect of diluent (nitrogen and/or argon) is extremely small.

Petersen et al. (2009) [9] measured ignition delay timings for propane and air mixtures within the temperature range of 870–1000 K and at a reflected shock pressure of 30 atm. The measured data were compared with the data of Cadman et al. (2000) [7], and they are in a good agreement. It was also shown that the reactions involving CH_3O, CH_3O_2, and $CH_3 + O_2/HO_2$ are important in reproducing the correct kinetic behavior.

Propane ignition delay was studied and modeled behind the reflected shock waves by Kim and Shin (2001) [10] at high temperatures ranging from 1350 to 1800 K, pressure range of 0.75–1.57 bar and at equivalence ratios of 0.5, 0.75, 1.0 and 2.0. An empirical correlation between ignition delay time and the concentrations of fuel-oxygen was proposed.

Healy et al. (2008) [2] studied methane-propane mixture and methane-ethane-propane mixture using a shock tube over a wide range of compressed gas pressures (10, 20 and 30 atm), gas temperatures (770–1580 K) and at equivalence ratios ranging from 0.3 to 3.0. They developed a kinetic model using HCT (hydrodynamics, chemistry and transport) program by Lund and Chase (1995) [11] and the model can mimic the ignition delay at various equivalence ratios and pressures.

Other shock tube investigations for propane ignition delay measurements are Lam et al. (2011) [12] work at reflecting shock wave pressures from 6 to 60 atm, temperatures from 980 to 1400 K, and at lean fuel–air mixture condition; Petersen et al. (2007) [13] work at pressures from 5 to 31 atm, temperatures from 1042 to 1585 K, and at equivalence ratios ranging from 0.5 to 3; Zhukov et al. (2005) [32] work at pressures ranging from 2 to 500 atm, temperature range of 880–1500 K and at lean fuel-air mixture conditions; and Penyazkov et al. (2005) [14] at pressure range of 2–20 atm, temperature ranges of 1000–1750 K, and equivalence ratios of 0.5, 1 and 2.0.

1.2. Propane studies using jet stirred and flow reactors

Dagaut et al. (1987) [15] studied propane oxidation at high temperatures of 900–1200 K, pressures of 1–10 atm and at equivalence ratios of 0.15–4.0 using jet stirred reactor. They developed a mechanism for oxidation of propane and compared the results with jet stirred reactor and available shock tube data from literature.

Propane oxidation was studied by Cathonnet et al. (1981) [16] at pressure between 1 and 6 bar, temperature near 1000 K and fuel-air ratios of 0.05–25 in laminar flow quartz reactor. Major products (C_2H_4, C_3H_6, CH_4, H_2, CO, C_2H_6) were sampled in this work. It was shown that the initial oxygen concentration has very little influence on ethene and propene yields, but has a significant effect on methane yields.

Shock tube and reactor simulations were performed by Westbrook and Pitz (1984) [17] for propane at pressure range of 1–15 atm, and temperature range of 1000–1700 K. New rate expressions were developed for a number of reactions of propane, propene, and intermediate hydrocarbon species. The mechanism also predicted correctly the laminar flame properties of propane and propene, and the detonation properties of propane.

Oxidation of propane was studied by Hoffman et al. (1991) [18] using a chemical flow reactor for pressure of 3.6–10 atm, equivalence ratio of 0.3, and at low temperature near 850–900 K.

Major product species were measured including C_3H_6, C_2H_5, CO, CH_4, CO_2, H_2, CH_2O, CH_3CHO, C_3H_6O, and C_2H_5CHO. They showed experimentally that the fuel conversion increases by increasing the

pressure and temperature. In addition, the product distribution shifts in favor of C_3H_6 over C_2H_4 by increasing the pressure and decreasing the temperature. A new detailed kinetic model was also developed and validated versus the measured data.

Sabia et al. (2014) [19] measured ignition delay times for propane to oxygen ratios of 0.03–0.8 using tubular flow reactor at atmospheric pressure over temperature range of 850–1250 K. Several combustion regimes were identified as a function of these external parameters. Ignition delay times were evaluated, and they showed different levels of dependence on the system inlet temperature for intermediate to high temperatures.

Other propane studies using flow reactor are available in literatures such as Levitsky et al. (1984) [20] work at pressure range of 172–250 torr, temperature range of 586–658 K, and equivalence ratio of 1.0; Beerer and McDonell (2011) [3] work at pressure range of 7–15 atm, temperature range of 785–935 K, and fuel-air ratios 0.4 and 0.6; Chang et al. (1958) [21] work at atmospheric pressure, temperature range of 1000–1153 K, and equivalence ratio of 1.0; and Holton et al. (2010) [22] work at temperature range of 930–1140 K, equivalence ratio range of 0.5–1.25, and at atmospheric pressure.

1.3. Propane studies using rapid compression machine

Ignition delay measurements were studied by Healy et al., 2008 [2] using a twin-opposed piston RCM configuration for methane-propane and methane-ethane-propane mixtures at pressures of 10–35 bar, temperatures from 740 to 1580 K and equivalence ratios of 0.5–3.0. Gallagher et al., 2008 [23] used the same twin-opposed piston RCM to measure ignition delay for fuel–air mixture at compressed gas pressures of 21–37 atm, fuel–air ratios of 0.5–2.0, and compressed gas temperature of 680–970 K. A negative temperature coefficient regime was observed at high pressure and experiments were compared with a newly developed kinetic model in their work.

1.4. Conclusion from literature

There are very scarce data regarding the propane ignition delay at low to intermediate temperatures using RCM. In addition, to the best of our knowledge, there is no measurements of excited radicals at auto-ignition conditions. The purpose of this study is to provide ignition delay data and excited radicals intensity over a wide range of compressed pressures, compressed temperatures, and equivalence ratio using a rapid compression machine at the wide range of pressures, temperatures and equivalence ratios.

2. Experimental setup

Rapid compression machine is a device to simulate the compression stroke of a single engine cycle to study the autoignition process in a controllable environment. The mixture of the fuel and oxidizer is compressed in a short time to reach a high compressed pressure and temperature. The compression time can be changed in our RCM from 28 to 40 ms to change the compressed temperature slightly, within approximately 5 deg. Shorter compression time decreases the heat transfer during compression stroke, which results to a slightly higher compressed gas temperature. The present RCM is pneumatically driven and hydraulically stopped. Stroke of the machine varies from 7 to 10 in. and clearance is also adjustable. Compression ratio of up to 12.33 can be obtained in this machine. The creviced piston in the RCM, based on the optimized design [24], minimizes the flow vortex during compression. The desired compressed conditions can be reached by changing initial pressure, initial temperature, and the compression ratio of RCM. The RCM has several heating bands to increase the temperature of the walls and test mixture to desired temperature. The test mixtures, as shown in Table 1, were heated inside the combustion chamber using controlled heaters to change the initial mixture temperature, and hence mimic