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## Experimental Studies of Laminar Burning Velocity and Autoignition of Hydrocarbon Fuels

Haoran Hu, Lawrence A. Kennedy
The Ohio State University

James C. Keck
Massachusetts Institute of Technology

#### Abstract

A constant volume bomb was used to determine laminar burning velocity of fuels at high pressure and temperature. Autoignition may occur during the laminar burning process at high temperature and pressure. Effects of diluent gas on the laminar burning velocity and autoignition are also studied. Autoignition has negligible effect on laminar burning velocity of fuel-air mixtures, but will accelerate the combustion process of fuel-oxygen-argon mixtures.

#### Introduction:

The laminar burning velocity and autoignition of adiabatically compressed combustible charges at high temperature and pressure are practical important in the design and analysis of internal combustion engines. Laminar burning velocity is among the most fundamental properties characterizing the combustion of homogeneous fuel-air mixtures, and the understanding of autoignition is important for the study of knock in spark ignition engines and ignition delay of diesel engines [1,2].

Laminar burning velocity, defined as the rate of flame propagation relative to the unburned gas, has been studied extensively both in experimental measurements [3,4] and in theoretical modeling [5,6,7] at low pressures. Some information is available in the range of pressure and temperature encountered in internal combustion engines, but none in the range of autoignition of unburned charges.

Autoignition is the onset of combustion in a reactive charge without the introduction of an external ignition sources. It is now widely accepted that engine knock is a result of autoignition of the fuel-air charge as it compressed and heated adiabatically by the motion of piston and by the expansion of burned gas behind the advancing flame front. But some experiments equipped with sufficiently fast diagnostics show the development of strong shocks with wave speeds as high as 2000 m/s [8], and some researchers argued that the engine knocking combustion was due to flame speed acceleration with a final transition to detonation [9]. Because of lacking studies of the transition process from laminar burning process to autoignition process, the controversy over knocking mechanism will continue.

In response to these needs, this paper is concerned with experimental studies of the transition process from laminar burning to autoignition of homogeneous fuel-air and fuel oxygen/argon mixtures.

#### Experimental Facilities and Procedures

The spherical combustion bomb used to measure the burning velocity. The bomb was assembled from flanged 450 alloy steel halves fastened by 6 clamp bolts and sealed by a Viton O-ring. It had an inside diameter of 153 mm and a 19 mm wall thickness and was designed to withstand pressure up to 1000 atm. Nickel plating on all bomb surface provided corrosion resistance and a smooth glossy surface.

The bomb was located in a 400 mm x 400 mm x 460 mm glass-wool oven and heated by eight 250 w resistance heaters mounted on the oven wall and by one 1400 Q resistance heater wrapped around the flanges of the bomb. All of the heaters were operated by one on/off temperature controller. An oven fan mounted above the bomb assured temperature uniformity of the oven gases. Four iron-constantan thermocouples distributed within the oven used to measure temperature. The accuracy of an individual measurement was 1 K and the bomb temperature was uniform within this accuracy.

Mixtures were ignited by stainless steel electrodes extending to the center of the bomb. A conventional discharge ignition system with variable voltage and capacitance served to generate the spark. Pressures were measured with a Kistler Model 621B piezoelectric pressure transducer. Chemi-ionization associated with flame front was measured by ionization probes mounted flush with the wall at three different location. Data was recorded using both an oscilloscope and a computer equipped with a/D converter. The initial conditions of the combustible charges in the constant volume bomb are critical to insuring the experimental results reliable and repeatable. The state of combustible mixture just before ignition should be quiescent and homogenous in temperature and species concentration. Details of the filling procedure are discussed in Ref[10,11] Typical oscillogram of pressure and ionization current traces is shown in Fig. 1.3. for stoichiometric hexane/air mixture at an initial temperature of 473 K and various initial pressures. The upper record is a typical normal burning process. Compressed by the growing laminar flame front, the pressure of the unburned mixture increases smoothly until it encounters the wall where it goes through a maximum and then decays slowly due to heat loss from the burned gas to the wall. The ionization probe signals show a spread in the arrive time of different arrival flame front at wall of approximately 0.5 ms. This is typical and indicates spherical symmetry of the flame front to better than one percent. Laminar burning velocity was calculated based on measured pressure signal. The two lower records show typical autoignition of end gas prior to completion of normal burning, but only the pressure signal in the normal burning portion was used to determine the burning velocity at the specified initial conditions.

#### Data Analysis

The combustion process in a spherical combustion bomb with central ignition can be modelled as an unsteady one-dimensional problem by assuming that all parameters are only a function of radial distance from the center of the bomb. The basic assumption are discussed by Metghalchi and Keck[3]. Based on these assumptions, the volume and energy balance equations for the gas in the bomb can be formulated.

The laminar burning velocity can be defined as

$$S_{u} = \frac{M\frac{dx}{dP}\dot{P}(t)}{A_{f}/V_{u}}$$
(3)

(4)

where P(t) is the measured pressure, and dP/dx can be expressed as

$$\frac{dx}{dp} = \frac{V_{i}[1 - (1 - x)\frac{V_{u}}{V_{i}}(1 - \frac{\gamma_{b}}{\gamma_{u}}) + \frac{A}{V}(\gamma_{b}p\frac{d\delta}{dp} + \delta)]}{\gamma_{b}(R_{b}T_{b}^{o} - R_{u}T_{u}}$$

where  $V_i$  = initial specific volume of the mixture,  $\gamma$  = specific heat ratio, R=gas constant, b = adiabatic flame temperature, V = volume of the combustion bomb, M = mass of the gas in the bomb, A = area of the bomb wall, E = initial internal energy in the bomb,  $\delta$  = thermal boundary layer displacement thickness of the unburned gas next to the bomb wall, V = specific volume, V = specific internal energy, V = mass fraction burned. As stated earlier, a number of basic assumptions are made in deriving the above expressions. The errors introduced by these assumptions are not expected to be over 1 percent

#### Results

Four fuels, butane, propane, n-octane and isooctane, used in the experiments were tested individually in both fuel air and fuel oxygen/argon mixtures. A typical raw result is shown in figure 2.1 for comparison with the results of Metghalchi and Keck[12] at same experimental facility and initial conditions. Burning velocities are plotted as a function of temperature, and it shows that the measurements of laminar burning velocity are reproducible. To avoid random errors, two runs were made for most experimental conditions.

#### Stoichiometric Fuel-Air Mixtures

Burning velocities of stoichiometric fuel-air mixtures are shown in log-log plots as a function of a ratio of unburned gas temperature to a constant reference temperature. In order to test the effect of autoignition on laminar burning velocity, initial temperature and pressure are elevated until autoignition can be observed. By assuming that the preflame chemical reaction effect is negligible before the autoignition occurs, laminar burning velocities are calculated from the measured pressure signals. In the cases of autoignition, the pressure signal is truncated at the point of autoignition occurs.

Experimental results of stoichiometric butane-air mixture are plotted in Fig. 3. At the initial pressure  $P_1 = 10$  and 15 atm, autoignition and pressure oscillation are plotted as a function of unburned mixture temperature ratio to the linear relations

$$\log(S_u) = a + \beta * \log(\frac{T_u}{T_{ref}})$$
(5)

where  $T_{ref} = 550$  K, which is the medium in the interested temperature range. The best fit parameters  $\alpha$  and  $\beta$  are obtained The value of B is the temperature exponent and a depends on pressure and fuel type. Because our focus in this study is to discuss the relation between laminar burning process and autoignition, the linear relation will give simplest and best fit to the measured data for each run. Fig. 7.1 shows the laminar burning velocity of stoichiometric octane-air mixtures. Solid curves are least fits to the linear relation.

#### Stoichiometric Fuel-Oxygen-Argon Mixture

For the fuel-air test, the temperature range spanned was about a factor of 1.5. In order to achieve higher temperature and pressure, nitrogen was substituted with argon as diluent gas. The average specific heat ratio of stoichiometric fuel/air mixture is 1.31 and the average specific heat ratio of stoichiometric fuel/oxygen/argon is about 1.5. With the substitution of argon to nitrogen as diluent gas, the unburned temperature range was extended from 720 K to 810 K.

Fig. 3.2-3.3 show the experimental measurements of laminar burning velocity of butane/argon/oxygen mixture. For fixed initial pressure at 7 atm, initial temperature ranges from 400 K to 500 K. Solid curves are least square fit to the linear relations. Fitting parameters  $\alpha$  and  $\beta$  indicate the laminar burning velocity is a weak function of initial temperature.

The effect of changing initial pressure on the laminar burning velocity of was also examined. For the normal laminar process, the burning velocity decreases as the initial pressure increases. During the processes of autoignition, the slope of burning velocity increases as the increases of initial pressure and autoignition intensity.

Fig. 8.1 and Fig. 8.2 show Laminar burning velocity of stoichiometric iso-octane/argon/oxygen mixture at initial pressure of  $P_i = 3.0$  atm to 16.0 atm and at fixed initial temperature  $T_i = 440$  K. Solid curves are least square fit to the linear relation. As shown in Fig. 8.1, burning velocity decreased when the initial pressure was increased from 3 atm to 5 atm and no autoignition was observed during the test. The slope of plotted curves started to change when autoignition was observed. In Fig. 8.2, the burning velocity increases with the increases of initial pressure and autoignition intensity.

#### Discussion

Comparison with previous measurements will be discused in the presentation.

The study of autoignition effect on burning velocity has fundamental importance on the mechanism of knocking combustion in the internal combustion engines. As discussed in Ref [9, 15, 16], two theories for the mechanism of knocking combustion have been favored through the years. First, the homogeneous autoignition theory identifies knock with thermal runaway of the chemical reactions in the end gas; Second, Detonation theory assumes the spark ignited turbulent flame to accelerate to detonation.

The present study show that for the fuel-air mixture, the burning velocity is unchanged regardless the autoignition condition. As indicated in the oscillogram in Fig. 1.4, there exists small pressure disturbance just

before the rapid pressure rise, but it has negligible effect on the burning velocity prior the autoignition occurs. It suggests that the rapid pressure rise was generated by the rapid chemical energy release of chemical reaction in the end gas.

The test results of fuel/oxygen/argon mixtures indicate that burning velocity was influenced by conditions of autoignition. The burning velocity increases rapidly as the pressure and temperature approach to autoignition condition. As discussed in the previous section, the chemical reaction in the thermal boundary is important to the acceleration of burning velocity at high temperature and pressure. But the rapid chemical energy release in the end gas, the unburned mixture outside the thermal boundaries, determined the autoignition in the combustion process. Hu and Keck[10] has successfully simulated the autoignition limits by using the measured pressure signals and by assuming there existed an adiabatic core in the end gas. But to provide quantitative explanation of the burning velocity acceleration, detail chemical kinetic analysis in the thermal boundary layer is recommended.

#### Summary

Measurements of the burning velocity of fuel/air and fuel/oxygen/argon mixture over a wide range of temperatures and pressures of interest for internal combustion engines and burners have been made. Special attention has been paid to the effect of autoignition on burning velocity. The fuels studies were butane, pentane, hexane, heptane, octane, and isooctane. Laminar burning velocities could be fitted to both linear relation and power law relation.

Autoignition has negligible effect on the burning velocity of fuel/air mixtures. The fast speed oscillogram of pressure trace show that small pressure disturbance exists prior to the major pressure increase, but it has very small effect on the burning speed prior to the autoignition.

Autoignition has significant effect on the burning velocity of fuel/argon/oxygen mixture at high pressure and temperature. Burning velocity increases as the increase of initial pressure and autoignition intensity. One possible explanation is the preflame reaction in the preheated thermal boundary layer due to higher adiabatic flame temperature and high thermal capacity of fuel/argon/oxygen mixture. Further studies of the preheated thermal boundary layer is needed to provide quantitative explanation.

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