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Visualisation of propane autoignition in a turbulent flow reactor using OH* chemiluminescence imaging

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ABSTRACT

Autoignition of propane in air was visualised in a turbulent flow reactor using natural OH*-chemiluminescence imaging. The spatial and temporal development of autoignition kernels was studied in an optically accessible tubular section of the reactor. Kernel nucleation, movement and growth affected the location and movement of subsequent autoignition sites, and resulted in stagnation of the incoming flow and flashback. The autoignition delays of the reactants were measured under various conditions of temperature, pressure and equivalence ratio, relevant to micro gas turbines: Temperature $T = 803–903$ K, pressure $p = 0.4–0.6$ MPa, equivalence ratio $\phi = 0.2–0.6$, mass flow rate of reactants $m_r = 8–21$ g/s, with ignition delays τ between 191 and 498 ms. The effect of diluting the propane + air mixtures with CO_2 was investigated for mole fractions of $0 < X_{CO2} < 0.1$. An empirical correlation for the autoignition delays was developed for the aforementioned conditions, and values of activation energy were calculated on the basis of the experimental data. The ignition delay times were compared with the predictions of chemical kinetic models and experimental data previously reported in the literature.

1. Introduction

Modern micro gas turbines employing dry-low-emissions combustors rely on the formation of lean-premixed fuel + air mixtures in a premixing tube, prior to the flammable mixture entering the combustor. These gas mixtures spend a finite time mixing under turbulent conditions in the premixing tube, at conditions of elevated pressure and temperature, often above their autoignition temperature. If autoignition is to be avoided within the premixing tube, the residence time of the fuel + air mixtures within the premixer tube must be limited to below that of their autoignition delay time. Autoignition delays depend on temperature, pressure and equivalence ratio of the reactants. In micro gas turbines employing lean premixed fuel + air mixtures, combustion chambers typically operate with inlet-air temperatures of around 600–750 K, pressures between 0.4 and 0.6 MPa, and primary-zone equivalence ratios ranging from $\phi = 0.2$ to 1.

Autoignition studies on propane have been reported in the literature using a variety of techniques. These comprise experiments carried out in flow-reactors, rapid-compression machines and shock-tubes. Flow reactors are typically able to operate at long autoignition times (100–500 ms) and low temperatures (600–1150 K) over a wide range of pressures (0.1–3 MPa) [1]. Due to their similarity with gas-turbine premixer flows, in having a turbulent flow at controlled pressure, it has been argued that flow reactors provide the most accurate representation of the conditions encountered in gas turbine premixers [2,3]. The applicability of analytical kinetic models such as the plug-flow-reactor model (PFR) to flow-reactor autoignition data though, is limited by the ability of the flow-reactor to produce instantaneously premixed reactants and plug-flow conditions. Flow reactors inherently have finite reactant mixing times, variations in initial reactant stoichiometry, turbulent mixing and diffusion of species in axial direction, as well as radially varying velocity distributions, all of which represent violations of these conditions. A detailed discussion on how these shortcomings may be addressed for the purpose of comparison of plug-flow reactor experiments with kinetic models has been described in the Supplementary material to the article of Zhao et al. [4]. Experiments have demonstrated that despite these shortcomings, even flow reactors operating under undiluted and turbulent conditions can provide reliable autoignition data, and show fair agreement with chemical kinetics predictions [1].

Some of the earliest flow-reactor experiments on propane in which the mixing time was short in comparison with the ignition delay time were devised by Jackson and Brokaw [5]. The autoignition delay was estimated as the difference between fuel injection time and onset of ignition, as detected by a pressure sensor or photocell in a transient laminar flow of fuel and air. Ignition delay times were reported for propane in air, at atmospheric pressure, for temperatures ranging from $T = 795$ to 1020 K for equivalence

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ratios ranging from $\phi = 0.25$ to 100. Measurements were limited to ignition delays above 200 ms.

Lezberg [6] conducted further flow-reactor ignition delay measurements at atmospheric pressure. The technique employed a steady flow of fuel and air to estimate the ignition delay time as the critical residence time of the mixture within the reactor, at which autoignition just occurred. The autoignition delays were determined by assuming instantaneous mixing and plug-flow of the reactants, and resolved ignition delays as short as 8 ms.

Lefebvre et al. [3,7] carried out flow reactor experiments similar to those of Lezberg [6] in which the variation of pressure on autoignition delay was investigated. This work reported ignition delay times for propane at pressurised conditions relevant to gas turbine premixers ($\phi = 0.15–0.8$, $T = 819–1000$ K, $p = 0.1–1$ MPa, $\bar{u} = 7–27$ m/s, $\tau = 18–105$ ms).

A detailed comparison of a steady flow technique similar to that employed by Lezberg [6] and Lefebvre et al. [3] and a transient flow technique comparable to that of Jackson and Brokaw [5], was carried out by Beerer et al. [8] under turbulent conditions and at elevated pressures. The investigation demonstrated that agreement exists amongst the steady flow techniques listed above. The transient flow technique was deemed to have potential advantages over the steady flow technique, such as not requiring knowledge of the flow-field. But Beerer et al. [8] concluded that additional care in ensuring rapid fuel injection (within 15–30 ms) was considered necessary when using the transient flow technique, in order to produce results that were comparable to those of the steady flow technique. In this and a further publication [1], Beerer et al. reported ignition delays for propane at a pressure of 0.9 MPa, temperatures ranging from $T = 839–886$ K and an equivalence ratio of $\phi = 0.6$. Agreement between the experimental data and chemical kinetic models, namely that developed by Petersen et al. [9] was reported.

Holton et al. [10] carried out transient autoignition experiments on propane in a flow-reactor at atmospheric pressure. The experiments were carried out for slightly higher temperatures than previous experiments, ranging from $T = 930$ to 1140 K, and at equivalence ratios of $\phi = 0.5$ and $\phi = 1$. The results showed reasonable agreement with the shock-tube measurements reported by Kim et al. [11], and with the ignition delay times predicted by the kinetic model of Petersen et al. [9].

An alternative experimental method of determining autoignition delay times using flow reactors is the use of rapid compression machines (RCMs). RCMs aim to raise the pressure and temperature of the reactants within a short time (e.g. 17 ms [12]) by reduction of the reactor volume to a smaller value, which is then held constant as reactions proceed. The technique allows heat release to be calculated from sensible rise in pressure, but has the disadvantage that the reactions take place at varying pressures. The technique also suffers from heat transfer losses of the reactants to the walls, making measurements of autoignition delay times in excess of 100 ms subject to increased inaccuracy [12].

Gallagher et al. [12] studied the autoignition of propane at temperatures in the range of 680–970 K, pressures of 2.1, 2.7 and 3.7 MPa, and equivalence ratios of 0.5, 1.0 and 2.0. The study used mixtures of propane and oxygen, diluted with nitrogen, argon or mixtures thereof. A negative temperature coefficient (NTC) region, in which autoignition occurred faster for reactants at lower temperatures than for the same reactants at higher temperatures, was reported between 730 and 850 K, at pressures of 2.7 and 3.7 MPa. Disagreement was reported between the autoignition delays determined in this study, and previously shock-tubes experiments carried out at similar low-temperature and high pressure conditions. The authors noted that shock-tube data may be prone to large experimental error at autoignition delay times longer than a few milliseconds.

The most suitable experimental technique for determining autoignition delays at higher temperatures ($T > 1250$) and ignition delays lower than a few milliseconds is likely to be the shock tube method. In this technique, a premixed quantity of reactants is heated by compression of a passing shock wave, ensuring short compression times, and, at short autoignition delays, a homogeneous mixture. At ignition delay times longer than a few milliseconds however, and particularly at low temperatures and high pressures, the accuracy of autoignition data obtained by means of shock tubes has been put into question. It has been shown [13–15] that ignition time measurements in shock-tubes taking longer than a few milliseconds may incur significant pressure and temperature increases due to chemical reactions prior to the main ignition event, and that the common assumptions of a shock tube behaving as a constant volume and internal energy reactor are not appropriate in these cases.

Burcat et al. [16] performed shock tube experiments using diluted propane + oxygen mixtures for temperatures of around 1250–1600 K and pressures of 0.2–1.0 MPa. The reported autoignition delay times ranged between 12 and 600 μs, and showed linear behaviour of $\log\tau$ versus 1/T within this range of conditions. Cadman et al. [17] carried out shock tube experiments at lower temperatures of 850–1000 K and at higher pressures between 0.5–4.0 MPa, at which autoignition times as long as 6 ms were recorded. The results showed significant changes in activation energy with respect to the data of Burcat et al. [16] and failed to be reproducible using existing chemical kinetic mechanisms, despite efforts to adapt their chemistry [14]. Herzler et al. [18] carried out experiments of propane-air mixtures at temperatures of 900–1300 K, 1–3 MPa and equivalence ratio of 0.5. Their experiments confirmed the findings of Cadman et al. [17] which showed a decrease in activation energy around 1050 K. The ignition delay times measured Herzler et al. [18] extended up to about 7 ms, and were shown by Aul et al. [15] to incur significant pressure and temperature increases due to chemical reactions prior to the main ignition event.

The autoignition studies in the literature provide only sparse description of the ignition location, and the spatial and temporal development of premixed autoignition flames in flow reactors, and of their dynamic behaviour. Those currently provided are mostly qualitative in nature. Lezberg [6] gave a verbal description of autoignition flames and their locations, noting the occurrence of blow-off and intermittent flashing of the flames. Further to this, steady-state photographs of diffusion flames, which were ignited by autoignition and stabilised at the fuel injector orifices, were provided in this study. Mullins [19] described discrete flame phenomena for the autoignition of liquid hydrocarbon fuels at different temperatures and locations within a flow reactor and noted that steady-state photographs of the flames were taken.

It has been reported that autoignition flames observed in flow-reactors were inherently unstable, and tended to cause flashback shortly after ignition. Mullins [20] observed that strong oscillations of the flames existed, and that the position of the autoignition flame front was ill-defined. Mullins [20] and Lefebvre et al. [3] both noted intermittent flashes of flames ahead of the steady autoignition flame front, which were visible when the flame was located downstream of the exit of the flow-reactor duct. Lefebvre et al. [3] further reported that flashback occurred when the autoignition flame entered the straight section of the flow-reactor duct. Beerer et al. [8] stated that it was impossible to establish a steady autoignition flame within the flow-reactor, due to flashback occurring immediately after ignition.

Given the limited availability of spatial and temporal descriptions of autoignition in premixed flow-reactors, the current study aimed to provide an account of the nucleation sites and the spatial