



Ignition delay times, laminar flame speeds, and mechanism validation for natural gas/hydrogen blends at elevated pressures



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ABSTRACT

New experimental ignition delay time data measured in both a shock tube and in a rapid compression machine were taken to determine the increase in reactivity due to the addition of hydrogen to mixtures of methane and natural gas. Test conditions were determined using a statistical design of experiments approach which allows the experimenter to probe a wide range of variable factors with a comparatively low number of experimental trials. Experiments were performed at 1, 10, and 30 atm in the temperature range 850–1800 K, at equivalence ratios of 0.3, 0.5, and 1.0 and with dilutions ranging from 72% to 90% by volume. Pure methane- and hydrogen-fueled mixtures were prepared in addition to two synthetic 'natural gas'-fueled mixtures comprising methane, ethane, propane, *n*-butane and *n*-pentane, one comprising 81.25/10/5/2.5/1.25% while the other consisted of 62.5/20/10/5/2.5% C₁/C₂/C₃/C₄/C₅ components to encompass a wide range of possible natural gas compositions. A heated, constant-volume combustion vessel was also utilized to experimentally determine laminar flame speed for the same baseline range of fuels. In this test, a parametric sweep of equivalence ratio, 0.7–1.3, was conducted at each condition, and the hydrogen content was varied from 50% to 90% by volume. The initial temperature and pressure varied from 300 to 450 K and 1 to 5 atm, respectively. Flame speed experiments conducted above atmospheric pressure utilized a 1:6 oxygen-to-helium ratio to curb the hydrodynamic and thermal instabilities that arise when conducting laminar flame speed experiments. All experiments were simulated using a detailed chemical kinetic model. Overall good agreement is observed between the simulations and the experimental results. A discussion of the important reactions promoting and inhibiting reactivity is included.

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1. Introduction

Fuel flexibility, increasing efficiency, and reduction in harmful emissions pose ever increasing challenges to the power generation industry. Due to increasing energy costs, dwindling traditional feedstocks and the simultaneous demand for cleaner energy in a power generation market competitive for efficiency and flexibility, the importance of alternative fuels sources is increasing. Energy sources such as coal gas, gases from industrial processes like coke manufacturing, biomass gasification and energy storage via hydrogen electrolysis have become potential alternative energy sources for the gas turbine industry. These gases are typically comprised of hydrogen, syngas (H₂/CO mixtures), and short-chain hydrocarbons generally classified as natural gas. Interest in hydrogen as a renewable energy source for use in gas turbines, fuel cells, and as

a transportation fuel has increased in recent years, as it is considered energy dense (on a mass basis) and environmentally friendly.

New routes to hydrogen formation have also led to increased attention; these include steam reformation of hydrocarbons, hydrogen generation via electrolysis of water, and coal/biomass gasification. A growing number of technologies are now harnessing the power of hydrogen to achieve the objective of reducing emissions and increasing fuel flexibility, including integrated gasification combined cycle power plants with carbon capture and storage and I.C. engines. All of this technological progress has made hydrogen fuel particularly interesting to the gas turbine industry. While operating gas turbines with pure hydrogen fuels poses some difficulties and requires significantly re-designed combustion systems and turbomachinery, the addition of hydrogen to natural gas mixtures is expected to alter the combustion properties only slightly [1], leading to a reduction in carbon monoxide, carbon dioxide and NO_x emissions from power generation gas turbine plants.

Even though natural gas is being widely used in the gas turbine industry, it does have some unfavorable combustion characteristics,

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such as a high flammability limit on the lean side and generally low reactivity which limits the operability to richer regimes. These limitations can be improved by the addition of hydrogen which can significantly lower the flammability limit of natural gas mixtures and extend the operability of natural gas turbines to leaner burning regimes [2]. Hydrogen can be mixed with natural gas and/or methane in concentrations of up to approximately 70% to assist the complete combustion of mixtures and reduce emissions. Previous studies have shown that, at up to approximately 50% hydrogen content by volume, the effect on fuel behavior is slight [1,3].

To implement mixtures of these fuels in gas turbines, detailed chemical kinetic mechanisms describing their combustion properties must be developed and validated against a wide range of experimental data for use in design-relevant simulations. In this study, a rapid compression machine (RCM), shock tube, and spherical flame have been employed to study the oxidation of natural gas/hydrogen blends over gas turbine-relevant conditions. Two natural gas mixtures have been studied to better reflect the varying composition of natural gas blends containing greater amounts of higher-order hydrocarbons derived from sources such as coal used in the power generation industry. The study of quaternary natural gas mixtures reflects better the real natural gas blends used in the gas turbine industry as opposed to their single- and binary-component counterparts, which have traditionally been utilized as natural gas surrogates.

Natural gas blends of interest to the gas turbine industry have been characterized previously. Healy et al. [4–9] studied quaternary natural gas mixtures in the temperature range 630–1550 K, in the pressure range of 10–30 bar and developed a detailed chemical kinetic mechanism for natural gas mixtures. Natural gas/hydrogen blends have also come under investigation recently. Park et al. [10] observed a reduction in reactivity when hydrocarbons such as propane or *n*-butane replaced methane in hydrogen/methane and syngas/methane flame mixtures. This reactivity reduction was determined to be a result of the depletion of hydrogen radicals through their consumption in reactions with ethylene (C_2H_4) and ethyl (C_2H_5) radicals. Reactivity also decreased due to the increased concentrations of methyl ($\dot{C}H_3$) radicals, which consumed further hydrogen atoms through the reactions $CH_4 + \dot{H} = \dot{C}H_3 + H_2$ and $CH_3 + \dot{H}(+M) = CH_4(+M)$, lowering the overall rate of the main chain branching reaction $\dot{H} + O_2 = \dot{O} + \dot{O}H$ [10]. Gersen et al. [11] reported ignition delay times of methane/hydrogen mixtures recorded in a rapid compression machine (RCM) at high pressure (10–70 bar) and intermediate temperature (950–1060 K). They observed that for hydrogen mole fractions above 50% there is a significant decrease in the ignition delay time compared to hydrogen concentrations below 20%, where the effect was slight.

Gersen et al. [11] also observed an increase in global activation energy with higher hydrogen content in the mixture, by reason of the differences in activation energy (E_a) between the two pure fuels, finding that at higher temperatures, the ignition delay time is more greatly reduced by the addition of hydrogen to natural gas mixtures when compared to lower temperatures. Gersen et al. [11] attributed this effect to the increasing importance of the $\dot{H} + O_2 = \dot{O} + \dot{O}H$ and $H_2 + \dot{O}H = \dot{H} + H_2O$ reactions at higher temperatures. Recently, Gersen et al. [12] expanded their study to include the effect of carbon monoxide on methane, hydrogen, and binary mixtures of the two fuels at high pressures (20–80 bar) and a lean equivalence ratio ($\phi = 0.5$). They found that for $CH_4/H_2/CO$ mixtures there was no inhibiting effect of CO addition compared to binary methane/hydrogen mixtures [12].

Crossley et al. [13] observed the effect of adding ethane, propane, *n*-butane, and *n*-pentane fractions to methane fuel. They determined that the addition of larger hydrocarbons led to a significant reduction in ignition delay time, and this effect was

determined to be due to the faster kinetics of the higher hydrocarbon fuels. de Vries and Petersen [14] measured undiluted natural-gas-based mixtures combining methane with ethane, propane, *n*-butane, *n*-pentane, and hydrogen at gas turbine relevant conditions in a shock tube. The results obtained showed a definite decrease in activation energy at lower temperatures and higher pressures as seen in the work of Gersen et al. [11]. Yu et al. [15] and Huang et al. [16] studied methane/hydrogen and propane/hydrogen, and methane/ethane/propane/hydrogen laminar burning velocities, respectively. They determined that an increase in hydrogen mole fraction in any case leads to an increase in laminar burning velocities. Few data have been published on hydrogen/hydrocarbon blend ignition delay times, and to the author's knowledge, no hydrogen/quaternary natural gas blend ignition delay times at gas turbine relevant conditions have been published previously.

While the flame speed of the main component of natural gas, methane, has been extensively studied in the literature [17,18], the laminar flame speeds of methane and natural gas blends with high hydrogen content at elevated temperatures and pressures remains to be studied. Because the flame speed of blended fuels cannot be obtained by linear combination of each blended fuel constituent, modeling work has been done to predict the flame speeds of methane and hydrogen blends by Chen et al. [19]. Correlations for similar fuel blends such as ethane and hydrogen mixtures have been studied by Wu et al. [20]. Flame speeds of butane–air mixtures with hydrogen addition have been experimentally determined in a study by Tang et al. [21], and Yu et al. [15] conducted methane flame speed experiments at atmospheric conditions with a small amount of hydrogen addition. Hu et al. [22] studied the entire range of methane and hydrogen blends at room temperature and pressure. No relevant laminar flame speed data at elevated temperatures and pressures were found in the literature.

As a result of the few fundamental kinetic studies available on methane and natural gas mixtures with hydrogen, the intent of this work was to provide a comprehensive set of ignition delay time and laminar flame speed data over a range of fuel blends at conditions of relevance to gas turbine engines. These data were used to compare with a modern, detailed chemical kinetic mechanism. By combining both RCM and shock-tube capabilities, a wide range of temperatures were examined for the measured ignition delay times. Details of the experimental apparatuses are provided first, namely the RCM, shock-tube, and constant-volume flame speed vessel, followed by an overview of the chemical kinetic mechanism. A comprehensive section detailing the results of the experiments in comparison with the model predictions is then provided. Further mechanism validation against archival data and a detailed sensitivity analysis are then presented.

2. Experimental setup

2.1. NUIG rapid compression machine

Experiments were performed in the rapid compression machine (RCM) facility at the National University of Ireland Galway (NUIG). The NUIG RCM has a horizontally opposed, twin-piston design that has been described in previous studies [23,24]. Compression times below 16 ms are achieved using this set up. The diameter of the reaction chamber is 38.2 mm. The chamber and sleeves are pre-heated to ensure a homogeneous temperature, and this heating can be carried out up to 160 °C. With the addition of creviced pistons, boundary layer effects are negligible, and the reacting core is temperature homogeneous and adiabatic. By varying the initial temperature, initial pressure, diluent gas composition, and volumetric compression ratio a wide temperature range can be investigated.

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