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## Influence of steam dilution on the ignition of hydrogen, syngas and natural gas blends at elevated pressures

Nicola Donohoe<sup>a</sup>, Karl A. Heufer<sup>a</sup>, Christopher J. Aul<sup>b</sup>, Eric L. Petersen<sup>b</sup>, Gilles Bourque<sup>c</sup>, Robert Gordon<sup>c</sup>, Henry J. Curran<sup>a,\*</sup>

<sup>a</sup> Combustion Chemistry Centre, NUIG, University Road, Galway, Ireland

<sup>b</sup> Department of Mechanical Engineering, Texas A&M University, College Station, TX, USA

<sup>c</sup> Rolls-Royce Canada, Montreal, Canada

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### ABSTRACT

This paper presents the influence of steam dilution on the autoignition behaviour of hydrogen, carbon monoxide, syngas, methane, and natural gas mixtures under gas turbine-relevant conditions. Rapid compression machine experiments were performed for fuel/air mixtures at equivalence ratios of 0.5, 1.0, and 2.0, in the temperature range 895-1140 K for the H<sub>2</sub> and CO mixtures and 730-1060 K for the natural gas mixtures and at pressures of 10 and 30 bar. Shock-tube experiments were performed for CH<sub>4</sub>-O<sub>2</sub>-Argon mixtures with and without H<sub>2</sub>O addition, highly diluted in argon (98% by vol.). The parameters were varied using an L9 Taguchi matrix for equivalence ratios of 0.5, 1.0, and 2.0; pressures of 1.6, 11, and 30 atm; and water contents of 0%, 10%, and 30% of the fuel by volume. It was found that significant changes in the thermal properties of the majority of the reactivity, whereas no chemical effect of the steam addition was observed for the majority of the mixtures investigated. Only mixtures of pure carbon monoxide were strongly influenced by water addition. In this case, the presence of water in the mixture allows the formation of relatively reactive OH radicals which enhances the possible oxidation chemistry of carbon monoxide leading to a greater observed reactivity of CO in the presence of water.

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#### 1. Introduction

Gas turbines are playing an increasingly important role in the generation of electricity. Their simple design, fuel versatility, and overall efficiency has allowed for their widespread implementation in electrical power generation. Gas turbines are also tolerant of a wide range of fuels and are now functioning satisfactorily using natural gas [1], gasified coal [2], and renewable energy sources such as hydrogen and syngas [3]. Hydrogen is expected to play an increasingly important role as a clean energy carrier in industry. But, in the short-term the cheapest and the most viable option for producing hydrogen with low  $CO_2$  emissions is to use fossil fuels with Carbon Capture and Storage (CCS) [4]. This option has become increasingly important in power generation due to expanding awareness of environmental issues such as the production of SOx, NOx [5] and  $CO_2$  [6] and the continuing abundance of natural

E-mail address: henry.curran@nuigalway.ie (H.J. Curran). URL: http://c3.nuigalway.ie/ (H.J. Curran). gas and coal reserves [7]. In the last decade, power-generating combined-cycle power plants have increased in overall efficiency. Improvements have afforded an increase in thermal efficiency to about 60% [8], while NOx emissions have been reduced by an order of magnitude, to below 9 ppm (dry, at 15%  $O_2$ ) in some cases [9]. For the near future, there appears to be no alternative other than to use fossil fuels for large-scale power generation. Continuous research into renewable fuels and increasing turbine efficiency are needed.

Research into increasing gas turbine efficiency has yielded some promising technologies. The combined-cycle power plant is now well established and offers superior performance to any of the competing systems which are likely to be available in the medium term for large-scale power generation applications [10]. Integrated Gasification Combined Cycle (IGCC) power plants with Carbon Capture and Storage (CSS) have become increasingly attractive for notable greenhouse emissions reduction and the ability to use coal and syngas as well as natural gas as fuel sources [11]. IGCC is a type of power technology particularly favourable for carbon dioxide capture as the CO<sub>2</sub> can be removed at a convenient stage of the process where its partial pressure is high [11]. This approach is

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 $<sup>\</sup>ast$  Corresponding author at: Combustion Chemistry Centre, School of Chemistry, NUI Galway, Ireland.

attractive as coal feedstocks are expected to last longer than those of natural gas, and syngas is a renewable energy source. ASUs (Air Separation Units) also play a key role in improving the efficiency, availability, and operability of oxygen-fed IGCC power plants. The use of CCS can typically reduce power plant  $CO_2$  emissions by around 85–95% [12]. An optimal integration between the ASU and the balance of the plant, especially the gasifier and the gas turbine, has significant potential for enhancing the overall plant efficiency.

Plant efficiency can also be improved by the use of heat recovery systems. Energy recovery from the exhaust gases and the generation of steam from processes such as Steam Injected Gas Turbines (STIG) are a normal practice to boost power in many applications. This process is a natural development of steam injection in regenerative cycles, which consists of a gas turbine combined with a Heat Recovery Steam Generator (HRSG) [13]. The steam used for injection is produced from fresh water in a HRSG by cooling the hot exhaust gases from the gas turbine. The steam is then supplied before or in the combustion chamber or at the turbine inlet. The flue gases and steam expand in the turbine and thereafter heat new water. Steam injection has been shown to improve thermal efficiency and reduce NOx emissions. Air from the compressor and steam from the HRSG both receive fuel energy in the combustion chamber and both expand inside the same turbine to boost the power output of the turbine. The specific heat of superheated steam is almost double the value of air and the enthalpy of steam is higher than that of air at relevant temperatures [13]. Therefore, the STIG method is a very effective way to boost the net power output and increase the overall efficiency of gas turbines [14].

To investigate the effect of steam on the fuel combustion behaviour, studies need to be performed using various gas turbinerelated fuels with steam addition. Moist hydrogen experiments have been performed previously in rapid compression machines [15], compression ignition engines [16], and shock tubes [17]; moist methane and hydrogen have also been investigated in jetstirred reactors [18]. However, to the authors' knowledge, a broad and in-depth ignition delay time study of steam addition to hydrogen, syngas, and natural gas mixtures has not been investigated. Consequently the aim of this study was to investigate the influence of steam on the ignition of hydrogen/CO and natural gas mixtures under turbine-relevant conditions. Experiments in a rapid compression machine and in a shock tube and simulations with a recent  $C_0-C_5$  kinetic mechanism were used to analyse the effect of water addition on the autoignition behaviour of these mixtures.

This paper details the results of the experiment and modelling study to examine the effect of water addition on the ignition chemistry of the target syngas, methane, and natural gas fuel blends. Presented first are the descriptions of the rapid compression machine and shock-tube experiments and related procedures, along with the mixtures that were studied. A description of the kinetic model and calculation approach is then given, followed by the experimental results and discussion. This last section begins with a model prediction study, followed by the results of the hydrogen experiments, the methane experiments, the natural gas tests, and CO experiments.

#### 2. Experimental details

#### 2.1. Rapid compression machine

Experiments were performed in the rapid compression machine (RCM) facility at the National University of Ireland Galway (NUIG). This machine has a horizontally opposed, twin-piston design that has been described in previous studies [19,20]. The pistons are

tightly sealed inside two compression sleeves that adjoin the reaction chamber [21]. The symmetry of the device helps to reduce the aerodynamic effects inside the combustion chamber at the end of the compression process. By the opposed movement of two twin pistons, compression times below 16 ms are achieved. The diameter of the reaction chamber is 38.2 mm, and special care has been taken to ensure a homogenous pre-heating of the reaction chamber is possible up to 160 °C. A large drive chamber is positioned behind each piston and serves as a reservoir of compressed air which is used to drive the pistons in place prior to compression. An hydraulic locking mechanism holds the pistons in place prior to compression. An hydraulic stopping mechanism brings the pistons to a complete stop at the end of compression. Creviced piston heads have been designed to reduce the formation of roll up vortices and boundary layer effects. By using flat piston heads, the cooler gases from the wall mix with the adiabatic reacting core and results in a non-uniform temperature distribution. Since the chemistry is very sensitive to temperature this makes interpretations and modelling very problematic [21]. With the addition of creviced pistons, boundary layer effects are negligible, and the reacting core is temperature-homogeneous and adiabatic.

Gas mixtures have been prepared in separate stainless steel mixing vessels with a minimum mixing time of 2 h before use. Table 1 presents an overview of the mixtures investigated. In each case, the oxygen/diluents ratio was 21/79, and a mixture of 50% nitrogen and 50% argon was used as the diluent. Only in the case of mixtures C1 and C2 was 100% argon used in the experiment to reach higher temperatures. Each mixture was investigated at 10 and 30 bar. A list of all the experimental data taken over the course of this study can be found in the Supplementary material.

The ignition delay time ( $\tau$ ) for the RCM tests was defined as the time from the end of compression, taken as the time of peak compressed pressure, to the onset of ignition, Fig. 1. As well as reactive experiments, non-reactive experiments where the oxygen content was replaced by nitrogen were also performed for each condition. From the pressure profiles obtained from these experiments, volume profiles have been deduced assuming isentropic compression/expansion. These volume profiles are used in kinetic simulations to account for facility effects, i.e. the change in pressure and temperature during compression and after the end of compression due to heat loss effects. The validity of this methodology has been shown in previous studies [19,20].

Special care has been taken to obtain the correct pressure profiles in the non-reactive experiment. The pressure transducer (Kistler 603B) used in this study is not heat shock resistant, i.e. the rapid temperature increase of the gas in the reaction chamber during compression and the resulting heat loads to the transducer respectively lower the output signal of the transducer. To prevent this effect from influencing the accuracy of the pressure

Table 1Mixture compositions investigated in % molar volume,<br/>the natural gas mixture (NG) contains 81.25% CH<sub>4</sub>, 10%<br/>C<sub>2</sub>H<sub>6</sub>, 5% C<sub>3</sub>H<sub>8</sub>, 2.5% C<sub>4</sub>H<sub>10</sub>, and 1.25% C<sub>5</sub>H<sub>12</sub>, and the<br/>syngas mixture (Syn) contains 25% CO, 50% H<sub>2</sub>, and 25%<br/>H<sub>2</sub>O.

Mixture	Fuel composition
H1	100% H <sub>2</sub>
H2	70% H <sub>2</sub> + 30% H <sub>2</sub> O
N1	90% NG + 10% H <sub>2</sub> O
N2	70% NG + 30% H <sub>2</sub> O
N3	70% NG + 15% Syn + 15% H <sub>2</sub> O
C1	100% CO
C2	70% CO + 30% H <sub>2</sub> O
C3	35% H <sub>2</sub> + 35% CO + 30% H <sub>2</sub> O

Note:  $\varphi$  = 0.5, 1.0, 2.0; p = 10 and 30 bar.

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