

IGNITION DELAY TIME MEASUREMENTS AND VALIDATION OF REACTION MECHANISM FOR HYDROGEN AT GAS TURBINE RELEVANT CONDITIONS

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ABSTRACT

The development of fuel flexible gas turbine (GT) combustors which are capable of operating with natural gas and H₂-rich fuels in a safe and reliable manner with low emissions is currently of significant interest to the GT industry. Gasification processes of coal, waste or biogenic material not only lead to the production of synthetic liquid fuels relevant to mobility, but also to highly efficient energy conversion processes, such as the next generation of Integrated Gasification Combined Cycle (IGCC) plants. Knowing the fundamental combustion properties of hydrogen and syngas mixtures including ignition delay time and flame speed at GT relevant conditions is therefore a prerequisite for a proper combustor design.

Ignition delay of hydrogen rich fuels at GT relevant conditions have been studied in both a rapid compression machine (RCM) and a shock tube. This work is part of the subproject "Combustion" within the European project "Low Emission Gas Turbine Technology for Hydrogen-rich Syngas" (H₂-IGCC). Ignition delay times of undiluted hydrogen/oxygen mixtures as well as diluted mixtures with Argon or nitrogen were measured at pressures up to 32 bar, at equivalence ratios between $\Phi = 0.1-4.0$ and at inlet temperatures higher than 900 K.

The experimental results show that the ignition times for hydrogen mixtures are highly dependant on both temperature and pressure. The ignition delays measured in both the RCM and shock tube are in agreement with each other. Several kinetic mechanisms available in the literature were tested against the experimental results.

There predictions are in good agreement with measurements performed.

Keywords: ignition delay time, mechanism validation, hydrogen

INTRODUCTION

Low-carbon fuels are currently of a high interest to the gas turbine (GT) industry to be employed in a broad range of primary energy sources. Besides different natural gas qualities, the use of H₂-rich fuels from coal or biomass gasification is of great interest. Future stationary gas turbines must be increasingly fuel flexible and able to operate with highly-reactive fuels without any deterioration in safety, reliability, and low-emission performance in comparison to modern lean premixed combustion systems. Operating such systems with highly reactive fuels significantly affects combustion properties such as ignition, flame stability, and flashback (Lieuwen et al. 2008).

One crucial combustion property, the ignition delay time, can be measured in RCMs or in shock tubes; as it is performed in the Combustion sub-project of the EU-project H₂-IGCC. The overall objective of this project is to provide and demonstrate technical solutions which will allow the use of state-of-the-art highly efficient, reliable gas turbines in the next generation of Integrated Gasification Combined Cycle (IGCC) plants. The goal is to enable combustion of undiluted hydrogen-rich syngas with low NO_x emissions and also allow for high fuel

flexibility. The challenge is to operate a stable and controllable GT on hydrogen-rich syngas with emissions and processes similar to current state-of-the-art natural gas GT engines.

There have been a limited number of studies of hydrogen ignition at high pressure and low temperature using a RCM and these have mainly been performed with stoichiometric mixtures. The main work was conducted by Lee and Hochgreb (Lee and Hochgreb 1998) who studied the autoignition process of hydrogen above the second ignition limit over the pressure and temperature ranges of 6–40 bar and 950–1050 K, respectively, for a slightly diluted mixture of $H_2/O_2/Ar$ (2/1/5). They compared their experimental results with a physical model of their RCM which takes heat loss into account. A new rate constant was recommended for the reaction $HO_2 + H_2 = H_2O_2 + H$ in order for the model to more accurately represent the experimental results.

Mittal et al. (Mittal et al. 2006) studied the autoignition of hydrogen as part of a work on H_2/CO (syngas) mixtures. Their work was conducted over a temperature range of 950–1100 K and at compressed pressures of 15, 30 and 50 bar for a diluted mixture of $H_2/O_2/N_2/Ar$ (2/1/2.9/10.1). Their results were in agreement with those of Lee and Hochgreb and show that the ignition times decrease with increasing compressed pressure. Mittal et al. compared their experimental results with numerous chemical kinetic mechanisms and concluded that the Ó Conaire mechanism (Ó Conaire et al. 2004) was the most accurate.

The only study conducted under lean conditions in an RCM was realized by Walton et al. (Walton et al. 2007) also as part of a syngas study. Their results were obtained over a pressure range of 10–15 bar and at a temperature of approximately 1030 K for an undiluted mixture of $H_2/O_2/N_2$ at an equivalence ratio of 0.1 and 0.15. Their pure H_2 experiments were only conducted to compare with previous RCM ignition studies and are in quite good agreement with Lee and Hochgreb results even if they were not achieved under identical conditions.

The most recent RCM work was conducted by Gersen et al. (Gersen et al. 2008) as part of a study on the effect of hydrogen addition to methane ignition properties. Their temperature and pressure ranges were 950–1060 K and 16–52 bar respectively, for a slightly diluted stoichiometric mixture of $H_2/O_2/Ar$ (2/1/5). Ignition delays results were in good agreement with previous studies by Lee and Hochgreb and Mittal et al.

There are a few hydrogen ignition delay time studies in shock tubes at high pressures and low temperatures by Blumenthal et al. (Blumenthal et al. 1995), Wang et al., (Wang et al. 2003) Pang et al. (Pang et al. 2009) and Herzler and Naumann (Herzler and Naumann 2009) at pressures up to 50 bar and equivalence ratios of 0.5 to 2. The experimental results at long ignition delay times and the lowest temperatures cannot be reproduced by available literature mechanisms for hydrogen combustion. This

mechanisms show a much higher activation energy compared to the experiments. The same tendency can be observed for syngas ignition delay time studies in shock tubes, see Dryer and Chaos (Dryer and Chaos 2008). A good agreement of the experiments with the predictions of different mechanisms can be achieved by considering the temperature increase due to gas dynamic effects during the measurement time, see Pang et al. (Pang et al. 2009) and Herzler and Naumann (Herzler and Naumann 2009). Small temperature increases of less than 20 K during the measurement time lead to a very significant reduction in ignition delay times due to the very high activation energy of the hydrogen system at low temperatures. Another possibility reason for the disparity between experiments and simulations may be catalytic effects (Dryer and Chaos 2008).

EXPERIMENTAL SETUP, MEASURING TECHNIQUES

Experiments were conducted for a different set of mixtures in the RCM and in the shock tube. The mixtures compositions are presented in Table 1. Different mixtures compositions are used in order to achieve different temperature range and reduce the heat loss in the RCM experiments. The dilution is used for safety reason in order to limit the heat and pressure release of mixtures with high hydrogen content and also helps avoid pre-ignition phenomena.

Table 1: Hydrogen mixtures tested

Device	Diluent				ϕ
	H_2	O_2	N_2	Ar	
RCM	0.7	1.0	0.752	3.008	0.35
	0.7	1.0	3.76		0.35
	1.0	1.0	0.752	3.008	0.5
	1.0	1.0	3.76		0.5
	1.0	1.0	13.4	13.4	0.5
Shock Tube	1.0	1.0	26.8		0.5
	0.2	1.0		23.6	0.1
	0.2	1.0		3.76	0.1
	1.0	1.0		26.8	0.5
	2.0	1.0		30.8	1.0
	8.0	1.0		54.8	4.0

Rapid compression machine (RCM)

The study carried out in NUI Galway is based on rapid compression machine experiments. This machine is a horizontally-opposed twin piston device which has been widely described in previous publications (Brett et al. 2001; Gallagher et al. 2008).

The symmetry of the system helps to reduce the aerodynamics inside the combustion chamber and the use of creviced pistons generates a homogeneous temperature field at the end of the compression stroke (Würmel and Simmie 2005). The piston heads have been adapted to

include a uniquely shaped crevice that captures the piston corner vortex, thereby maximizing the homogeneity of the temperature field at full compression. As a result, both the temperature field and mixture distribution inside the combustion chamber are optimized for homogeneity at full compression.

Thermodynamic conditions of pressure and temperature relevant to those in gas turbines are reproduced in the RCM. These conditions are achieved by a very rapid adiabatic compression process that takes 16–17 ms to complete. After the compression stroke, the pistons are held in place until the fuel auto-ignites. The thermodynamic conditions are therefore kept constant in the combustion chamber until ignition occurs.

Experiments were carried out over the temperature range 900–1060 K and compressed gas pressures of 8, 16 and 32 bar. Different compressed gas pressures were achieved by changing the initial pressure. The range of compressed temperatures was achieved by changing the initial temperature and by changing the composition of the diluents. The diluent is a mixture of inert gases of varying specific heat ratios, in this case nitrogen and argon. The ratio of the specific heat of nitrogen ($\gamma=1.28$) is lower than that of argon ($\gamma=1.66$) which results in a lower compressed gas temperature when nitrogen is used. For most of the experiments, the oxidizer portion consisted of 21% oxygen and 79% diluent. One diluted mixture (with a ratio of 1:5) has also been tested. Fuel-oxidizer mixtures were prepared manometrically in stainless steel tanks. All gases used for the experiments had a purity of 99.9 % or higher.

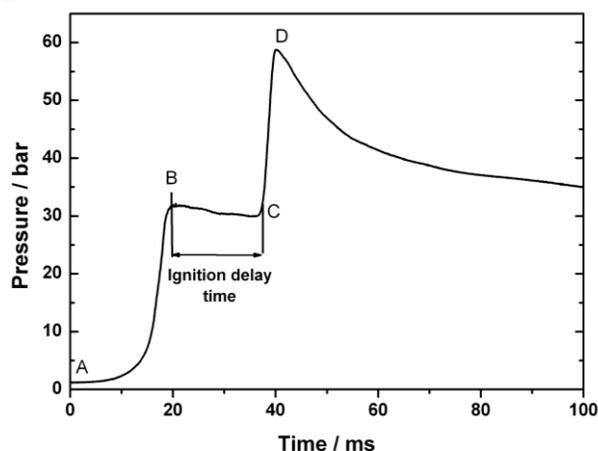


Figure 1: Pressure profile (Experimental conditions: $H_2/O_2/N_2/Ar$: 0.7/1/0.94/2.82 Initial Pressure: 543 mbar, Initial Temperature: 331 K)

During the auto-ignition experiments, the piston position and the pressure profile are recorded and saved on a digital oscilloscope, see Figure 1. The pressure profile, recorded using a pressure transducer (Kistler 603B), provides (i) the compression time (from A to B), (ii) the ignition delay time (from B to C), defined as the time between the end of the compression and the maximum of the heat release rate, and (iii) the maximum pressure (point D).

Shock Tube

The experiments were carried out in a high pressure shock tube with an internal diameter of 98.2 mm. It is divided using aluminium diaphragms into a driver section of 5.18 m and a driven section of 11.12 m in length. The driven section can be evacuated to pressures below 10^{-6} mbar using a turbomolecular pump. Gas mixtures were prepared manometrically in a stainless steel storage cylinder, which is evacuated using a separate turbomolecular pump to pressures below 10^{-6} mbar. The shock speed was measured over three 20 cm intervals using four piezo-electric pressure gauges. The temperature and pressure behind the reflected shock wave were computed from the measured incident shock speed and the speed attenuation using a one-dimensional shock model. The estimated uncertainty in reflected shock temperature is less than ± 15 K in the temperature range of our measurements.

The ignition was observed by measuring pressure profiles with piezo-electric gauges (PCB[®] 113A24 and Kistler[®] 603B) located at a distance of 1 cm to the end flange. The PCB[®] gauge was shielded by 1 mm of polyimide to reduce heat transfer. Also, the OH* emission at 308 nm at the same position was selected using a narrow band pass filter (FWHM = 5 nm) and measured with a photomultiplier. All ignition delay times shown were determined by measuring the time difference between the initiation of the system by the reflected shock wave and the occurrence of the OH* maximum as this allows comparisons by simulations. An observation time window of up to 6.5 ms can be achieved depending on the reflected shock temperature.

The temperature increase during the experiments due to gas dynamic effects was considered by using MPFR (Multiple Plug Flow Reactor) - CHEMKIN II, a programme developed at DLR Stuttgart to account for gas dynamic effects causing pressure and temperature variations decoupled from the effects of heat release combined with pressure relaxation effects along the shock propagation direction due to the shock tube's 'open end' configuration. Thus, the simulation assumes for a time period of typically 25 μ s or shorter depending on the heat release ($\Delta T \leq 0.5\%$), a PFR with constant pressure conditions and takes into account the propagation of the pressure increase by heat release within a PFR-time step along the shock propagation direction. The correction of the gas dynamic effects is based on the pressure profiles measured for mixtures with similar acoustic properties but with no heat release due to chemical reactivity. The temperature profiles are then calculated by applying adiabatic and isentropic conditions. These temperature profiles can be used instead of constant initial temperatures T_5 for the simulation of experimental profiles with different chemical mechanisms.

RESULTS AND DISCUSSION

RCM

The experimental results were compared with modeling results based on various chemical kinetic mechanisms. The results are presented in Figure 2 and 3. As RCM experiments always involve heat loss processes during both the compression phase and the constant volume phase, the model is based on an experimental pressure trace obtained for a mixture which has the same thermodynamic properties as the reactive mixture (O_2 is replaced by N_2) in order to take this phenomenon into account. Thus, this pressure trace is used to calculate the volume profile which is used as an input to CHEMKIN (Kee et al. 1990). Heat losses are considered to be an adiabatic expansion process as discussed previously by (Mittal et al. 2007; Gallagher et al. 2008) in their simulations.

The experimental compressed temperature is calculated from the initial temperature, T_{i_s} and pressure, p_{i_s} the compressed pressure, p_c and the mixture composition. The compressed pressure is defined as the pressure reached at the end of the compression (point B in Figure 2). The compressed temperature is calculated assuming an adiabatic compression with the Gaseq code (Morley) which considers the dependence of γ to the mixture composition and the temperature in the following formula (eq. 1) and assumes frozen chemistry during the compression process.

$$\ln\left(\frac{p_c}{p_i}\right) = \int_{T_i}^{T_c} \frac{\gamma}{\gamma - 1} \frac{dT}{T} \quad \text{eq. 1}$$

The auto ignition phenomenon was studied by changing the initial temperature of the mixture. Thus, no ignition was recorded below 900 K for these experimental conditions. It can be seen in Figs. 3 and 4 that the temperature range investigated in the RCM is quite narrow because of the high sensitivity of H_2 to the compressed temperature. The ignition delay decreases with both the compressed temperature and pressure. Moreover, the composition of the diluent has a large impact on ignition times. This is due to the reduction of heat loss when only N_2 is used as a diluent. In this case, the temperature profile is higher than in the 80%Ar / 20% N_2 case, which explains a shorter ignition delay due to a higher reactivity. However, for shorter ignition delays (less than 30 ms), the heat loss effect becomes less predominant and the pure N_2 results are in agreement with the other ones. This is especially noticeable for the results obtained at $\Phi = 0.5$ and a compressed pressure of 32 bar. The ignition delays measured at a higher equivalence ratio, $\Phi = 0.5$, (Figure 3) are shorter than those at $\Phi = 0.35$ (Figure 2).

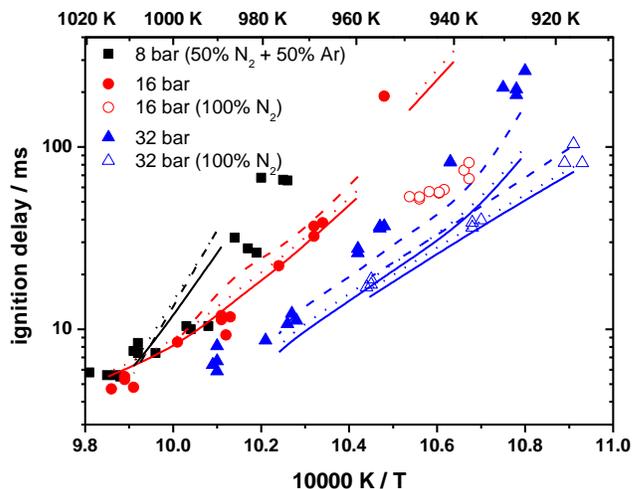


Figure 2: Effect of pressure on ignition delay for a mixture of H_2/O_2 /diluent (0.71/3.76) and comparison of the experimental results (Symbols) with the numerical results (lines: solid Ó Conaire updated, dashed: Ó Conaire, dotted: Li et al.)

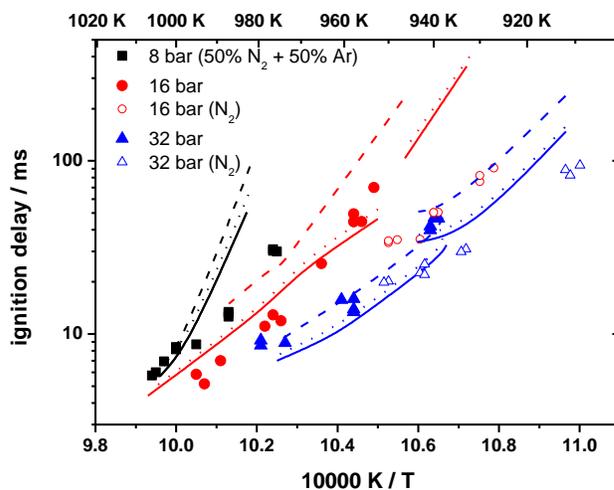


Figure 3: Effect of pressure on ignition delay for a mixture of H_2/O_2 /diluent (1/1/3.76) and comparison of the experimental results (Symbols) with the numerical results (lines solid Ó Conaire updated, dashed: Ó Conaire, dotted: Li et al.)

The experimental results have been compared to three different kinetic mechanisms: Ó Conaire (Ó Conaire et al. 2004), an updated version of Ó Conaire and Li (Li et al. 2007). The models show quite a good agreement with the experimental data except for the 16 bar experiments which were conducted in nitrogen. In this case, all the models produce ignition delays which are up to five times longer. The Ó Conaire and its updated version perform well for $\Phi = 0.35$ at 8 bar and at 32 bar when N_2 is used as a diluent. The performance of the Li mechanism is very good at 32

bar but it does not perform as well at 16 bar especially at $\phi = 0.5$. All the models also have some trouble in reproducing the lowest temperature at 8 bar. For both equivalence ratios, no model was able to produce an ignition at the lowest temperature.

Shock tube

The experimental results together with simulations using two different kinetic mechanisms: an updated version of Ó Conaire (Ó Conaire et al. 2004) and Li (Li et al. 2007) are shown in Figs. 4–9.

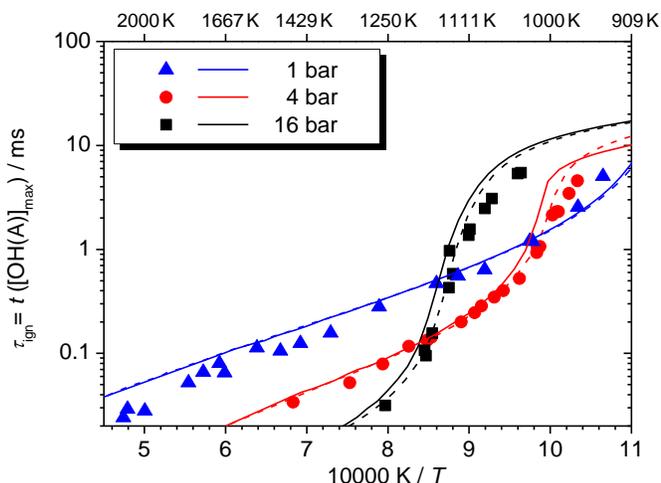


Figure 4: Effect of pressure on ignition delay for a mixture of $\text{H}_2/\text{O}_2/\text{Ar}$ (0.2/1/23.6) and comparison of the experimental results (Symbols) with the numerical results (lines: solid: Ó Conaire updated, dashed: Li et al.).

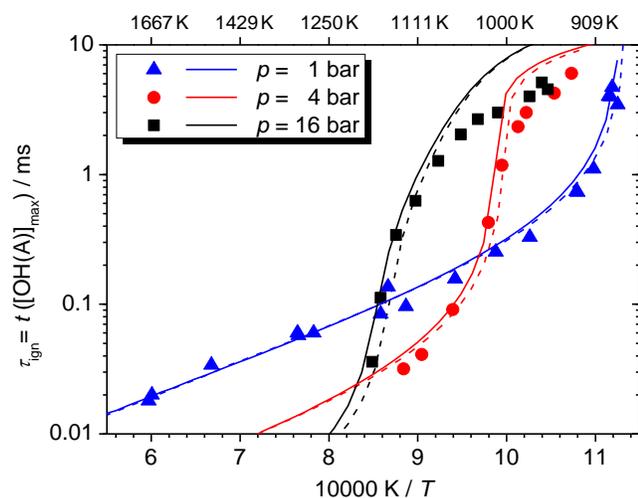


Figure 5: Effect of pressure on ignition delay for a mixture of $\text{H}_2/\text{O}_2/\text{Ar}$ (0.2/1/3.76) and comparison of the experimental results (Symbols) with the numerical results (lines: solid: Ó Conaire updated, dashed: Li et al.).

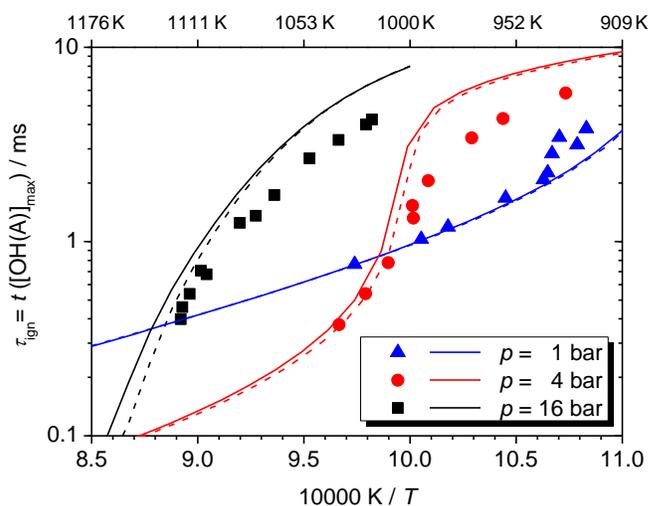


Figure 6: Effect of pressure on ignition delay for a mixture of $\text{H}_2/\text{O}_2/\text{Ar}$ (1/1/26.8) and comparison of the experimental results (Symbols) with the numerical results (lines: solid: Ó Conaire updated, dashed: Li et al.).

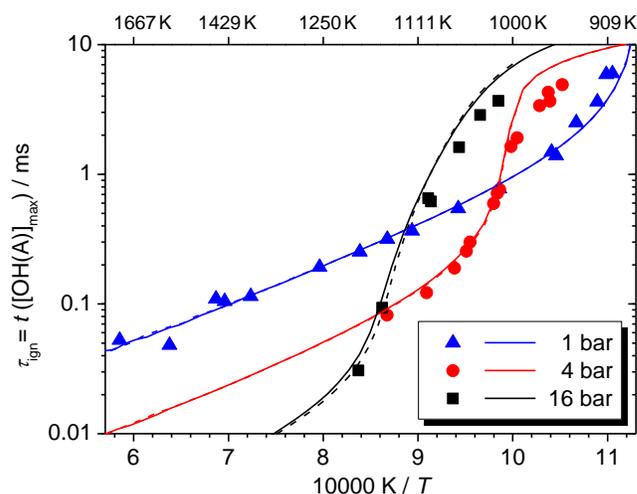


Figure 7: Effect of pressure on ignition delay for a mixture of $\text{H}_2/\text{O}_2/\text{Ar}$ (2/1/30.8) and comparison of the experimental results (Symbols) with the numerical results (lines: solid: Ó Conaire updated, dashed: Li et al.).

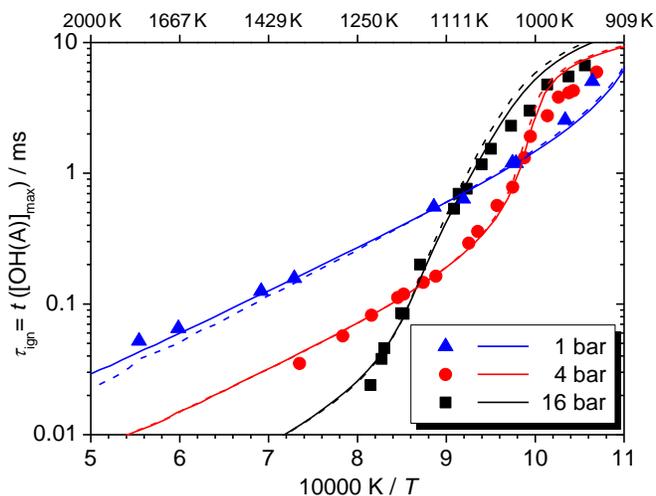


Figure 8: Effect of pressure on ignition delay for a mixture of $\text{H}_2/\text{O}_2/\text{Ar}$ (8/1/54.8) and comparison of the experimental results (Symbols) with the numerical results (lines: solid: Ó Conaire updated, dashed: Li et al.)

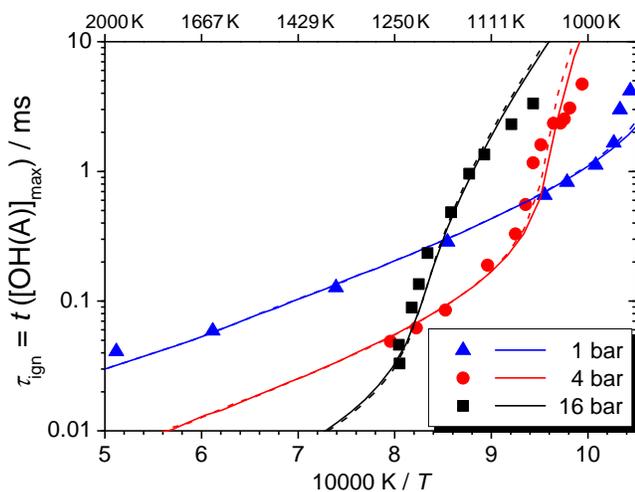
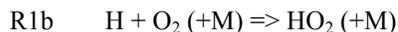


Figure 9: Effect of pressure on ignition delay for a mixture of $\text{H}_2/\text{O}_2/\text{N}_2$ (1/1/26.8) and comparison of the experimental results (Symbols) with the numerical results (lines: solid: Ó Conaire updated, dashed: Li et al.)

All measured ignition delay times show a very complex pressure dependence. There are intersections of the measurements at different pressures. At higher temperatures the ignition delay times at higher pressures are shorter whereas at lower temperatures the ignition at lower pressures is faster. This can be explained by the transition from the first to the second explosion limit. At higher temperatures the reaction:



is dominant. Higher pressures favour the reaction progress due to the higher absolute concentrations of the reactants. At lower temperatures the reaction:



becomes dominant. This reaction channel is pressure dependent and is therefore favoured by higher pressures. It inhibits chain branching via reaction R1a and thus leads to longer ignition delay times at higher pressures.

The measurements in the RCM are in the regime of the third explosion limit, where higher pressures favour shorter ignition delay times.

The different regimes can be seen using isothermal simulations of an undiluted $\text{H}_2 / \text{O}_2 / \text{Ar}$ mixture at 8, 16 and 32 bar, see Fig. 10. The shock tube measurements are in the region of the red box, the lowest temperatures represent the values with the highest activation energy, whereas the RCM measurement are in the region of the blue box due to the lower temperatures, higher pressures and undiluted conditions.

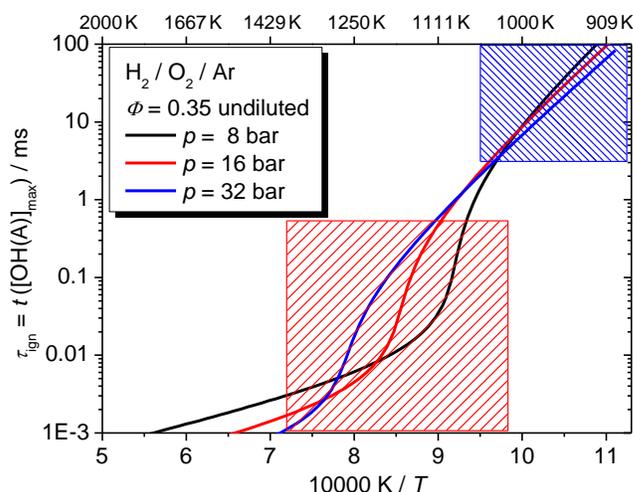


Figure 10: Simulated ignition delay times for a $\text{H}_2 / \text{O}_2 / \text{Ar}$ (0.7 / 1 / 3.76) mixture using the Li et al. mechanism.

The influence of reaction R1b is also very clearly shown by comparing the ignition delay times with Ar and N_2 as inert gas, see Fig. 11. The ignition delay times with N_2 as inert gas are significantly longer due to the higher collision efficiency of N_2 compared to Ar favouring reaction R1b. The ignition delay times of the 1 bar experiments at higher temperatures are identical for both inert gases because R1b is not significant in this temperature and pressure region. At lower temperatures the measured values in N_2 become clearly longer than in Ar due to the increasing importance of reaction R1b.

Comparing the measurements at different equivalence ratios it can be seen that the values at $\Phi = 0.5$ and 1.0 are

almost identical whereas the values at $\Phi = 0.1$ and 4.0 are also very similar but clearly higher than at $\Phi = 0.5$ and 1.0. The undiluted mixture at $\Phi = 0.1$ exhibits shorter ignition delay times compared to the diluted mixture.

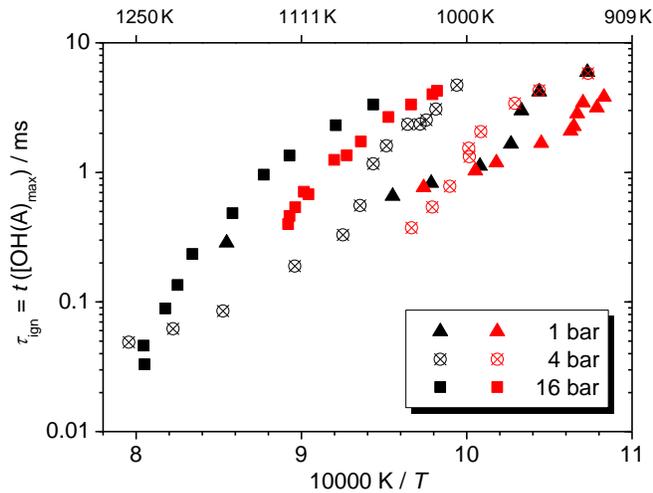


Figure 11: Dependence of the ignition delay times of a $H_2/O_2/Ar$ or N_2 (1/1/26.8) mixture on the inert gas. Red symbols: Ar, black symbols: N_2 .

By comparing the experimental results with simulations using different literature mechanisms it can be concluded that most of them are in good agreement with the experiments. The Li et al. and the updated version of Ó Conaire mechanism show a very good agreement for all measured values with the exception of the longest ignition delay times and lowest temperatures at $p = 4$ and 16 bar. In this region too long ignition delay times are predicted. The definition of the exact experimental conditions for this region is very difficult. The hydrogen system exhibits a very high activation energy at these temperatures and pressures, see the simulation without temperature correction in Fig. 12. Very small temperature differences lead to very significant changes in the ignition delay times. The temperature corrections which are calculated using the pressure profiles must therefore be very accurate. Another possible problem in this region is that there are 'weak ignition' effects leading to shorter measured ignition delay times. The most significant differences between measurements and simulations are observed for the undiluted mixture at $\Phi = 0.1$. They can be explained by the measured pressure profiles. Before ignition a strong pressure increase is observed analogous to measurements of Pang et al. see Fig. 13. This pressure and temperature increase cannot be explained by using homogenous kinetics. It is caused probably by ignition farther away from the end flange. Using a calculated temperature profile based on the measured pressure profile a very good agreement of simulations and experiments can be reached. The differences in the predictions of the Li et al. and the updated version of the Ó Conaire mechanism are quite small. Both are very well-suited for representing the

hydrogen chemistry in the validated temperature and pressure range.

The extension of the measurement range to lower temperatures and longer ignition delay times by the RCM experiments show that the observed deviations of our experiments to the simulations are caused by experimental problems in this region. The RCM experiments exhibit also a good agreement between experiments and simulations for lower temperatures. The mechanisms are therefore validated for the whole temperature, pressure and equivalence ratio range of the measurements.

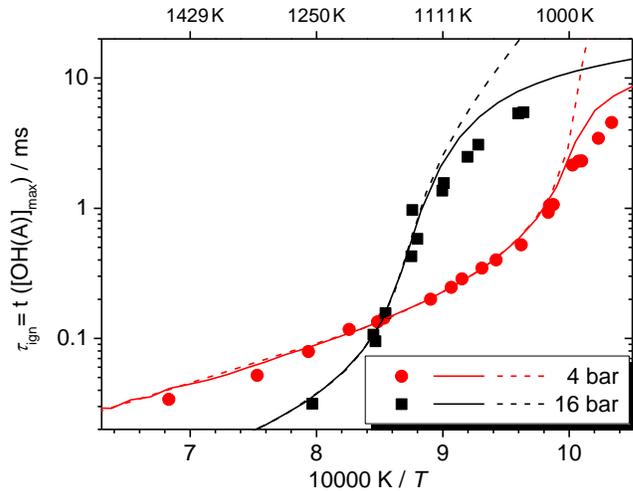


Figure 12: Effect of pressure on ignition delay for a mixture of $H_2/O_2/Ar$ (0.2/1/23.6) and comparison of the experimental results (Symbols) with simulations with the Li mechanism using constant temperature conditions (dashed lines) and temperature profiles based on measured pressure profiles (solid lines).

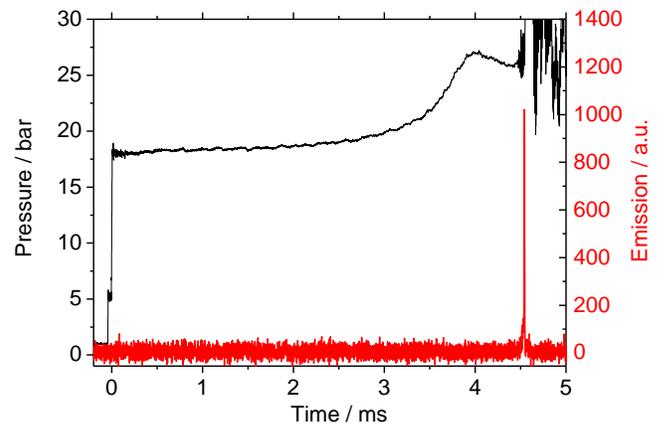


Figure 13: Experimental pressure and OH^* -emission profiles of a undiluted $H_2/O_2/Ar$ (0.2/1/3.76) - mixture at $p_{5,0} = 17.35$ bar and $T_{5,0} = 956$ K. Black line: pressure, red line: OH^* -emission.

Comparison

The ignition times measured in both the shock tube and the RCM have been compared at 16 bar for the diluted $\Phi = 0.5$ experiments (Fig. 14). Firstly, the shock tube results show that ignition delay times are faster when Ar is used as a diluent (black points) compared to N_2 (blue points). This is due to the higher efficiency for N_2 relative to Ar in the reaction $H + O_2 + (M) = HO_2 + (M)$ and in $H_2O_2 + (M) = OH + OH + (M)$.

At low temperature the RCM results show a higher temperature dependence than those obtained in the shock tube. There are two phenomena which explain this difference: (i) heat loss in the RCM reduces the temperature and tends to inhibit the reactivity for long ignition times; (ii) gas dynamic effects (mentioned before) increase the temperature inside the shock tube resulting in shorter measured ignition times.

The Ó Conaire mechanism has been used to calculate the ignition delay over the whole temperature range. The dashed black curve is a simulation assuming an increasing temperature in the shock tube due to gas dynamics effects. The solid black (Ar diluent) green (50% Ar, 50% N_2 diluent) and blue (N_2 diluent) curves are adiabatic, constant pressure simulations. The red curve is a simulation of the low temperature RCM data, which takes into account non-reactive pressure profiles recorded in the RCM. Calculated ignition times show good agreement with the RCM results. In all cases the ignition time is calculated based on the maximum in OH or OH^* concentration.

The shock tube simulations (the solid blue curve) are in good agreement with the experimental data performed using N_2 as diluent at temperatures of 1070 K and higher but below 1070 K the simulation is slower than the experiment due to experimental gas dynamic effects which seem to increase at lower temperatures.

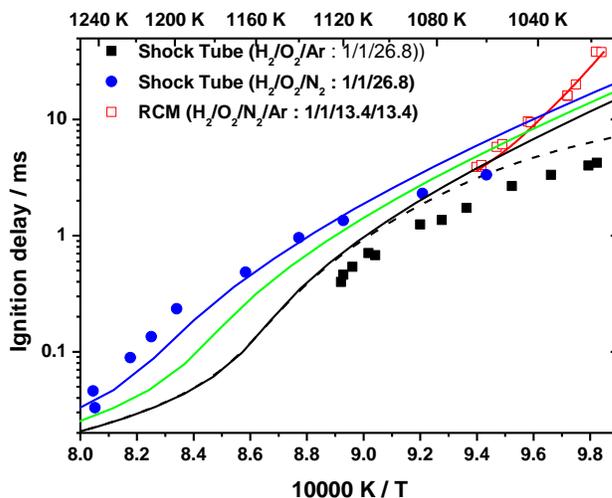


Figure 14: Comparison of the ignition delay in the shock tube and the RCM at a compressed pressure of 16 bar and $\Phi = 0.5$ (symbols: measurements, lines calculated ignition delay with the updated Ó Conaire mechanism based on $[OH]_{\max}$ or $[OH]^*_{\max}$: red: RCM, solid black, green and blue: constant pressure adiabatic simulation for Ar, 50% Ar / 50% N_2 and N_2 as diluent, dashed black: shock tube assuming increasing temperature for Ar as diluent)

SUMMARY AND CONCLUSIONS

The present work has studied the ignition delay of hydrogen over a wide range of experimental conditions relevant to gas turbines in both a shock tube and an RCM. The ignition delay shows a high sensitivity to the compressed pressure and temperature. Experimental results have been compared to different mechanisms from the literature and most of them are in good agreement. Finally, the comparison of the results measured on both experimental devices shows a good agreement.

It is expected that these results will act as a basis for the future studies of H_2/CO mixture oxidation in the RCM and the shock tube under conditions relevant to gas turbines.

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