



Parametric investigation of water loading on heavily carbonaceous syngases



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ABSTRACT

An outwardly propagating spherical flame was used to characterise the influence of water loading on the pre-mixed combustion of an applied high CO/H₂ ratio syngas fuel blend (converter gas). A nonlinear extrapolative technique was used to obtain values of laminar flame speed for combustion with air, for varying temperature, pressure and equivalence ratio. With increased attention given to the accurate measurement of laminar flame speed, a concerted effort was made to quantify experimental uncertainty, and a detailed methodology is presented. Change in relative humidity was shown to have a substantial impact on laminar flame speed for the syngas, increasing measured values by up to 70% from the driest cases. This observed increase results from the dissociative influence of H₂O addition, and enhancement in the formation of chain carriers that catalyse CO oxidation, increasing net heat release rate. In addition to relative humidity, the decoupled influences of initial temperature and pressure were investigated parametrically; holding the mass ratios of fuel and H₂O constant for a step change in condition. Temperature rise was shown to enhance H₂O induced acceleration, with greater relative change in heat release rate for a corresponding drop in flame temperature, and the opposite effect observed for increased pressure. The effect of water addition was shown to be non-monotonic, with flame speed reduction achieved at the highest water loadings for the hottest tests, and discussed as a function of initial CO/H₂ ratio. Attention was given to the dominant reaction kinetics, with the performance of several published reaction mechanisms evaluated against experimental data using CHEMKIN-PRO; with flame speed consistently overpredicted when H₂O was added to the mixture. A modified reaction mechanism is presented for the humidified combustion of high CO/H₂ mixtures, changing the rate parameters of two chain branching reactions to give higher relative indeterminate H₂O formation, and a reduction in OH carriers. Results obtained using the modified mechanism demonstrate improved agreement with all experimental data presented here and from a previous study, including changes in H₂O concentration at elevated temperatures and pressures. The results also highlight relative humidity as a potential source of error in the experimental measurement of u_L , significant for fuels comprising large CO fractions, but also potentially for other gaseous fuels, emphasising that relative humidity should be carefully considered when comparing experimental data.

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1. Introduction

The influence of water content on syngas combustion is becoming an increasingly prominent area of research, as engineers strive for the application of alternative fuels in cleaner, more efficient and complex technologies [1–5]. Central to this development is the compound chemical influence that water can have catalysing CO oxidation, in competition with a lowering of adiabatic flame temperature: Direct

formation of CO₂ from CO is slow due to a high activation energy, and the presence of hydrogen facilitates chain branching OH formation, changing the dominant path for oxidation [6]. Water addition can therefore provide a non-monotonic influence on premixed flame propagation, with lower concentrations catalysing oxidation through disassociation, whilst higher concentrations will eventually dilute the reaction and temperature as the accelerative influence is lost. The work presented employs a heavily carbonaceous syngas (Basic Oxygen converter gas, which typically comprises 50–80% CO, 10–18% CO₂, 1–3% H₂ in a balance of N₂ (%vol) [7]) as a case study for detailed parametric investigation of this behaviour. It represents a practical fuel which is increasingly used throughout the industrialised world [7].

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Nomenclature

A	area
A_a	pre-exponential factor
A_A, B_A, C_A	Antoine equation coefficients
E_a	activation energy
k_f	rate constant
Ka_{mid}	Karlovitz number normalised midpoint stretch rate
L_b	Markstein length
Le	Lewis number
m_f	mass of fuel
m_o	mass of oxidiser
Ma_{jin}	Markstein number representing slope of flame speed on stretch
P	initial pressure
r_{sch}	schlieren flame radius
R_{Sn}	uncertainty in regression
S_u	unstretched flame speed
S_n	stretched flame speed
t	time
T	initial temperature
T_{AD}	adiabatic flame temperature
u_L	laminar flame speed
U_{uL}	uncertainty in laminar flame speed
U_{Su}	uncertainty in unstretched flame speed
v_i	independent variable
x_f	molar fuel fraction
X	normalised condition
y_i	fixed error in variable
α	flame stretch-rate
η	mass Ratio
θ	gaseous Impurity
ρ_b	density of the burned gas
ρ_u	density of the unburned gas
σ_{Su}	standard deviation in S_u
ϕ	equivalence ratio

Subscripts

A, B, C mass ratio identifiers

1.1. Aim

The laminar flame speed (u_L), sometimes referred to as laminar burning velocity, or burning rate, represents one of the most important fundamental physiochemical properties of a fuel mixture. It is often used as input to detailed combustion models or to validate chemical reaction mechanisms [8]. Previous work [9] undertaken by the authors and presented in Fig. 1, quantified the non-monotonic changes in u_L possible from variation in relative humidity, as a function of H_2/CO ratio for another heavily diluted carbonaceous by-product fuel (note that 0.75 g $H_2O \approx 72\%$ RH at 303 K, and model predictions were generated using different reaction mechanisms in CHEMKIN-PRO). Due to the diluted nature of this fuel blend, detailed investigation was problematic given the small changes in flame speed realised from water addition, relative to experimental uncertainty.

The aim of the presented work was to experimentally investigate the influence of water content with the combustion of a practical fuel mixture comprising a higher CO fraction. Variation in air/fuel ratio was analysed, in addition to parametric studies of the influence of initial temperature and pressure. Particular attention was afforded to changes in intermediate chemistry and reactant thermo-diffusivity through the analysis of several chemical models and quantification of the burned gas Markstein Length (L_b). Experimental data were attained using an outwardly propagating spherical flame configuration,

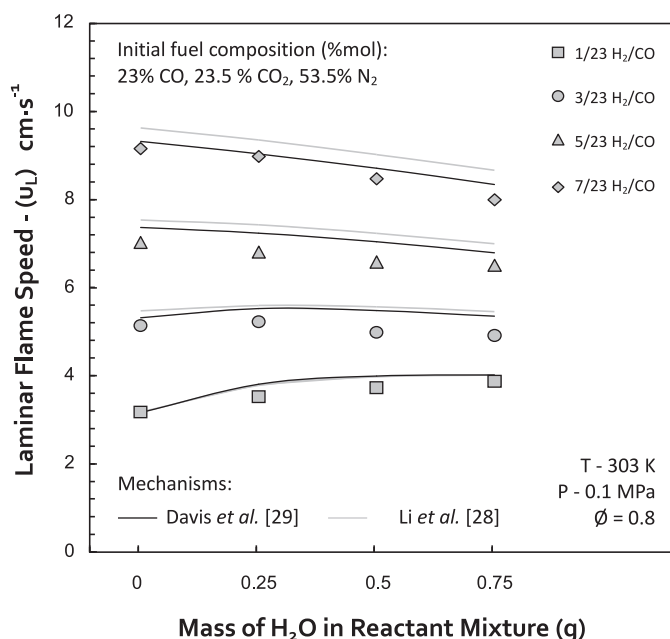


Fig. 1. Experimental (symbols) and modelled changes in laminar flame speed of humidified blast furnace gas mixtures, for combustion with air [9].

within a constant-volume combustion bomb (CVCB) which has been fully characterised previously [10].

Results were used to adapt and develop an existing chemical reaction mechanism, improving the comparative performance in modelling humidified syngas mixtures. A detailed methodology for estimating experimental uncertainty was also employed in this study, and is presented alongside the results.

2. Experimental setup

2.1. System overview

A schematic overview of the experimental setup employed is shown in Fig. 2. The CVCB has an internal volume of approximately 34 L (internal diameter: 260 mm), designed to allow for a sufficiently long experimental time window in the constant pressure region of flame expansion [10]. A PID control system regulates the ambient reactant temperature (T). Four diametrically opposed 100 mm quartz viewing windows facilitate high-speed imaging of flame propagation, by employing the widely used schlieren optical technique [10–14]. Images were captured by a CCD high-speed camera (Photron FAST-CAM APX-RS ($\pm 0.05\%$)) at a rate of 5,000 fps. The system allowed for a spatial resolution of ~ 0.14 mm per pixel, with propagation rates calculated by bespoke software employing commercially available edge-detection algorithms.

H_2O was injected into the evacuated system through a self-sealing septa (see Section 2.2), with the liquid mass measured on a high-precision balance (Mettler Toledo AE50 (± 0.2 mg), resolution 0.1 mg). Gaseous fuel and air were then introduced into the chamber using multiple mass flow controllers (Bronkhorst mini CORI-FLOW devices ($\pm 0.5\%$ Rd)). The total mass required for each reactant was pre-programmed, enabling both controllers to supply batch controlled mass up to the desired value. Mass fractions were calculated as a function of initial pressure (P) equivalence ratio (ϕ), temperature, and water loading. Partial pressures were recorded using a 0–2000 mbar sensor (BOC Edwards ASG ($\pm 0.2\%$ FS), resolution 0.1 mbar), with a real-time instrument controller readout, and were used to compare with equivalent calculated values. This was a secondary control to ensure that the correct quantities of liquid H_2O mass had fully

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