Fuel 189 (2017) 66–78

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

Experimental observations on the influence of hydrogen atoms diffusion on laminar and turbulent premixed burning velocities

A.A. Burluka [a], R.G. Gaughan [b], J.F. Griffiths [c], C. Mandilas [a,*], C.G.W. Sheppard [a], R. Woolley [d]

[a] School of Mechanical Engineering, The University of Leeds, Leeds LS2 9JT, UK
[b] ExxonMobil Research and Engineering Company, Paulsboro Technical Centre, 600 Billingsport Road, Paulsboro, NJ 08066, USA
[c] School of Chemistry, The University of Leeds, Leeds LS2 9JT, UK
[d] The University of Sheffield, Department of Mechanical Engineering, Mappin Street, S1 3JD, UK

ARTICLE INFO

Article history:
Received 24 May 2016
Received in revised form 17 October 2016
Accepted 18 October 2016
Available online 25 October 2016

Keywords:
Laminar flames
Turbulent flames
Burning velocity
Hydrogen combustion
Deuterium combustion
Isotope effect

ABSTRACT

Measurements of the laminar and turbulent burning velocity of premixed hydrogen–air, *n*-hexane–air and *n*-octane–air flames were made and compared to corresponding measurements of deuterium–air, *n*-hexane-d14–air and *n*-octane-d18–air flames performed at identical initial conditions. Experiments were conducted in a constant volume, optically accessed vessel, at elevated initial pressure and temperature of 0.5 MPa and 360 K, for a range of equivalence ratios. Burn rate data was determined via schlieren imaging of flames. It was found that the isotope effect accounted for an average reduction of 20% in the laminar burn rate of alkanes. Similarly, deuterium was measured to burn around 30% slower than hydrogen at the range of equivalence ratios explored. The isotope effect on burn rate was significantly reduced under turbulence. The difference between the turbulent burn rates of the deuterated alkanes and their normal alkane counterparts were measured to be approximately 10%. The difference between the turbulent burn rates of deuterium and hydrogen was even smaller. Nonetheless, the laminar burn rate ranking was maintained under turbulence for all fuels and conditions explored, thus suggesting a degree of influence of radical transport and chemistry under turbulent burning.

1. Introduction

The molecular structure of a fuel (i.e. length of chain, branching, bonding) is known to greatly influence the laminar burn rate [1–5]. Fuel structure in conjunction with mixture stoichiometry, pressure and temperature, govern the thermodynamics and chemical kinetics of combustion. One of the main driving forces of chemical kinetic contributions to the control of burning velocity is the radical pool at the flame front, with H atoms being the most important by virtue of their extremely high diffusivity and reactivity [4,5].

In two previous papers we investigated the effects of fuel structure on the laminar and turbulent burning velocities of gasoline components. We assessed:

(i) isomeric structure and bonding through experimental studies of seven different hydrocarbons containing six carbon atoms, over a wide range of fuel - air mixtures [6] and

(ii) chain length and molecular mass by reference to straight chain alkanes in the range C_5–C_8 [7].

Interpretation of the results was linked to the influence of H radicals at the flame front in controlling the burn rate.

The object of the present work was to understand the importance of the transport and kinetic effects of hydrogen radicals within laminar and turbulent premixed flames via experimental studies of burning velocity measurements of freely propagating flames. Hence, we have extended the earlier studies [6,7] through comparisons of laminar and turbulent velocity of *n*-hexane and *n*-octane (i.e. *n*-C_6H_{14} and *n*-C_8H_{18}) with those of their fully deuterated forms (i.e. *n*-C_6D_{14} and *n*-C_8D_{18}). In interpretation of the results, it is assumed that there are no qualitative differences in the kinetic mechanisms involved in flame propagation of the normal and deuterated fuels. In addition, we explore the H/D isotopic effect in its most influential guise, via an investigation of the laminar and turbulent burning velocities of H_2 and D_2.

The available literature on the isotopomoric effects in combustion is sparse. With the exception of a study of laminar flame propagation in acetylene and di-deuteroacetylene by Friedman and Burke [8], we are not aware of any other investigation of the lam-

* Corresponding author at: The Centre for Research and Technology, Hellas, Chemical Process & Energy Resources Institute, 3km Charilaou-Thermi Road, Thermi 57001, Greece.
E-mail address: mandilas@cperi.certh.gr (C. Mandilas).

http://dx.doi.org/10.1016/j.fuel.2016.10.088
0016-2361/

Nomenclature

Latin symbols

D	mass diffusivity (m^2/s)
k	reaction rate coefficient (m^3/mol s)
L	integral length scale of turbulence (m)
L_b	burnt Markstein length (m)
P_i	initial pressure (Pa)
r_u	cold flame mean flame radius (m)
T_{ad}	adiabatic flame temperature (K)
T_i	initial temperature (K)
S	laminar burning velocity sensitivity factor (–)
u'	turbulent root-mean-square velocity (m/s)
u_l	unstretched, one-dimensional laminar burning velocity (m/s)
u_n	stretched, entrainment laminar burning velocity (m/s)
u_{te}	entrainment turbulent burning velocity (m/s)

Greek symbols

α	thermal diffusivity (m^2/s)
α_{mix}	thermal diffusivity of mixture (m^2/s)
α	flame stretch rate (1/s)
δ_l	laminar flame thickness (m)
η	Kolmogorov length scale of turbulence (m)
ϕ	equivalence ratio (–)
ω	global reaction rate (m^3/mol s)^n

inar and turbulent burn rates of deuterated versus normal hydrocarbons, even though the former are occasionally used for tracing the origins of pollutants in flames, e.g. [9]. Moreover, although hydrogen burn rate data exist for a variety of conditions [e.g. 10–18] there is only very little information on the laminar burn rates of D_2 [19–21] and, to our knowledge, no comparisons between the turbulent burn rates of H_2-air and D_2-air flames.

2. Experimental apparatus and data processing

All measurements were performed in the Leeds MkII spherical bomb [22]. As in the previous studies for the examination of fuel structure and chain length effects on burn rate [6,7], measurements were performed at elevated temperature and pressure (360 K and 0.5 MPa), at which the premixed turbulent flames demonstrate behaviour similar to flames in spark-ignition engines [22]. Owing to the high cost of the deuterated fuels, the alkane tests were undertaken at only $\phi = 0.8$ and $\phi = 1.0$ for laminar conditions and at $\phi = 1.0$ under turbulent conditions. For the latter, the turbulence level was set at an rms velocity of $u' = 4$ m/s. The burn rates of H_2 and D_2 were examined for laminar and turbulent ($u' = 4$ m/s) conditions for $0.6 \leqslant \phi \leqslant 1.1$. The turbulence rms velocity of $u' = 4$ m/s chosen for this study was relevant to reciprocating engines, where u' near the top dead centre is about half the piston speed (e.g. $u' = 5$ m/s, for 75 mm stroke, at 4000 rpm [23]).

Premixed mixtures were prepared inside the fan-stirred vessel. Pre-calculated volumes of liquid fuels were injected into the vessel under vacuum conditions, using a gas tight syringe. For gaseous fuels, the mixture stoichiometry was controlled by measuring the partial pressure of the fuel injected into the bomb at atmospheric pressure. The bomb fans were continuously operated during mixture preparation to ensure full mixing and to assist heat transfer from the 2 kW electrical heater, positioned close to the walls of the vessel. For laminar studies, the fans were switched off for a period of 60 s, following mixture preparation, and before ignition. For turbulent studies the fans were maintained at the speed required to produce the desired rms turbulent velocity throughout the mixture preparation, ignition and combustion period. The pressure in the vessel before ignition was measured via an absolute pressure transducer (Druck PDCR-911) with a range of 0–0.7 MPa. Following spark discharge, the pressure rise in the vessel was monitored with a Kistler-701 piezoelectric pressure transducer, flush mounted on the side of the vessel. After each experiment the vessel was flushed several times with compressed air and then evacuated. Dry cylinder air was provided for the combustible mixture.

At least two laminar and five turbulent deflagrations were performed at each condition. Centrally ignited advancing flames were imaged via the schlieren method to the bomb window diameter of 150 mm, using a Photosonics Phantom Series 9 high speed digital camera. Laminar flames were recorded at 4000 frames/s with a resolution of 576 × 576 pixels. Turbulent flames were photographed at a rate of 9000 frames/s with a resolution of 384 × 384 pixels.

During post-processing of schlieren data, each flame image was converted from grayscale to black and white. White corresponded to the burned and black to the unburned region. The flame area was found by counting the number of white pixels. The burning velocity was then defined as the radius derivative with respect to time divided by the ratio of densities of the fresh mixture to that of the combustion products. Mean flame radius was determined as that of a circle encompassing the same area. Further information on the flame image processing procedure is given in [6,24]. Imaging data analysis to obtain laminar flame characteristics (burning velocity, stretch rate and Markstein lengths) and turbulent burn rates followed established methods, detailed [25,26] and widely used elsewhere [eg. 27–31].

3. Results

Presented in this section are measured laminar and turbulent burn rate results for normal and deuterated alkanes, hydrogen and deuterium. Experimental scatter for laminar deflagrations was at a maximum of 2% with respect to the coefficient of variance (COV) of the laminar burning velocity at any given flame radius. Turbulent deflagrations reported here exhibited an average scatter of circa 7–8% in COV of the turbulent burn rate at a given flame radius, which was similar to that reported in [32] and is typical of the magnitude of cycle-to-cycle variation of the burning rate in an SI engine [33].

In addition to data for the unstretched, one dimensional, laminar burning velocity, u_l, derived in accord with [26], the laminar results for the alkanes also include data for the stretched laminar burning velocities, u_n, at mean flame radii of 10 mm and 30 mm. Due to the comparable molar mass of *n*-hexane versus *n*-hexane-d14 and *n*-octane versus *n*-octane-d18, stretch rate effects were anticipated to be similar and, therefore, to not affect the trends observed for the laminar burn rate ratio of normal vs deuterated alkanes.

For H_2 and D_2 laminar deflagrations, hydrodynamic flame instabilities [34,35] occurred too early to apply the criteria for unstretched laminar burning velocity [26]. It was thus decided to present the schlieren-derived stretched burning velocities at mean flame radii of 10 mm and 30 mm to allow comparison to previous studies [13] and ensure exclusion of spark effects [36].