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Measurements of Flat-Flame Velocities of Diethyl Ether in Air

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Abstract

This study presents new adiabatic laminar burning velocities of diethyl ether in air, measured on a flat-flame burner using the heat flux method. The experimental pressure was 1 atm and temperatures of the fresh gas mixture ranged from 298 to 398 K. Flame velocities were recorded at equivalence ratios from 0.55 to 1.60, for which stabilization of the flame was possible. The maximum laminar burning velocity was found at an equivalence ratio of 1.10 or 1.15 at different temperatures. These results are compared with experimental and computational data reported in the literature. The data reported in this study deviate significantly from previous experimental results and are well-predicted by a previously reported chemical kinetic mechanism.

Keywords

Laminar burning velocity; Diethyl ether; DEE; High temperature; Combustion

1. Introduction

Oxygenated biofuels are important in today's world due to a decline in oil and fossil fuels and an increase in CO₂ emissions and atmospheric pollution. Furthermore, as the price of fossil-derived fuels rises, focus is being drawn towards bio-derived fuels, the most common of which is bio-ethanol. Bio-ethanol is used predominantly in the United States and Brazil, in mixtures with gasoline of 10 and 85 vol. %; however, bio-ethanol is produced primarily from food sources (corn, sugarcane, *etc.*) and its energy output is not ideal [1, 2]. Diethyl ether (DEE) is produced by dehydration of ethanol and is considered to be an excellent compression-ignition fuel [3]. It has a cetane number above 125 and a larger heating value than ethanol.

Davoud and Hinshelwood [4] studied the thermal decomposition of DEE with focus on the amount of acetaldehyde present at each stage of the reaction. Freeman, Danby and Hinshelwood [5-9] also studied the thermal decomposition of diethyl ether and focused on rate-pressure relations, effect of reaction conditions, nitric acid as an inhibitor, production of cyanides and production of ethanol. Waddington [10] investigated the gaseous oxidation of DEE at low temperatures and Laidler and McKenney [11,12] examined its pyrolysis from 833 to 893 K and 15 to 320 mmHg and the inhibition of pyrolysis by nitric acid from 833 to 913 K and 10 to 360 mmHg.

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Diethyl ether has previously been investigated as an additive with common fuels in use in different engines. Mack *et al.* [13] experimentally investigated the addition of DEE to ethanol in a homogenous charge compression ignition (HCCI) engine in order to find a fuel blend that will elongate the combustion event and reduce the autoignition temperature relative to ethanol. However, they did not observe these desired results due to the similar nature of DEE and ethanol. Mosbach *et al.* [14] numerically simulated this data and predicted that the addition of diethyl ether accelerated combustion due to the greater ignitability of DEE. Miller Jothi *et al.* [15] explored the use of DEE as a cetane number improver for use with liquefied petroleum gas (LPG) in a HCCI engine. Their findings show a reduction in nitric oxide (NO), particulate matter (PM) and smoke emissions and an increase in hydrocarbon (HC) and carbon monoxide (CO) emissions compared to diesel fuel. Sudheesh and Mallikarjuna [16] added diethyl ether to biogas in a HCCI engine in order to improve ignition. This addition resulted in higher brake thermal efficiency and lower HC, NO and smoke emissions, however, an increase in CO emissions was observed. Cinar *et al.* [17] studied the effects of adding DEE to diesel fuel in a HCCI direct-injection engine and found that nitrogen oxide (NO_x) and soot emissions decreased and CO and HC emissions increased simultaneously. Iranmanesh *et al.* added DEE to diesel [18] and biodiesel [19] in a diesel engine and found that NO_x emissions decreased with addition to both fuels. However, they found that HC emissions increased with addition to biodiesel and carbon dioxide (CO₂) emissions increased with addition to diesel.

Recently, Yasunaga *et al.* [20] studied the oxidation and pyrolysis of DEE at high temperatures behind reflected shock waves. Their results showed that the most important reactions in the combustion of DEE in a shock-tube are a four-centered elimination (reaction 1), C-O bond fission (reaction 2), and hydrogen abstraction to the secondary radical, C₂H₅OC₂H₄-s (reaction 3).

Agnew and Agnew [21] investigated the cool and two-stage flames of diethyl ether formed in flat-flame burners using infrared radiation techniques and observed emissions from excited formaldehyde in the range from 3200 to 5100 Angstroms. Griffiths and Inomata [22] studied the oscillatory cool flames of diethyl ether in air experimentally and numerically to investigate the low temperature oxidation of diethyl ether, which, they found, was associated with the hydroperoxide CH₃CH(OOH)OC₂H₅.

Burning velocities of DEE have been measured by Gibbs and Calcote [23] using a Bunsen burner flame cone and the apex-cone method. Di *et al.* [24] and Zhang *et al.* [25,26] used a spherically propagating flame in a cylindrical vessel to obtain laminar burning velocities and study the combustion characteristics of diethyl ether. However no results have been reported for DEE flame velocities using the heat flux method.

This study presents new adiabatic laminar burning velocities of diethyl ether in air, measured on a flat-flame burner using the heat flux method [27]. The experimental pressure was 1 atm and initial temperatures of the fresh gas mixture ranged from 298 to 398 K. These results are compared with the experimental data of Gibbs and Calcote [23] and Di *et al.* [24], and simulations using the mechanism of Yasunaga *et al.* [20].

2. Experimental Method

The heat flux method was first proposed by de Goeij *et al.* [27] in 1993. This method is beneficial because stretching of the flame does not occur and therefore stretch correction or extrapolation of the data are not necessary. This particular apparatus has previously been used for measuring laminar flame velocities of components of surrogate mixtures of natural gases [28].

In the flat-flame adiabatic burner, a brass burner plate, used to stabilize the un-stretched flame, is mounted on a plenum mixing chamber, Fig. 1. The burner plate is 2 mm thick and 30 mm in diameter. The plate is perforated with small holes of diameter 0.5 mm, with the pitch between them being 0.7 mm. Eight Type K thermocouples, of diameter 0.5 mm, are soldered to the plate surface at different distances and angles from the centre to the edge of the plate. The plenum chamber is encompassed by a thermostatic oil jacket, the temperature of which is set to the desired initial temperature of the unburned gas mixture. The circumference of the burner plate is heated with thermostatic oil set to 50 K above the temperature of the unburned gas mixture. In practice, if an initial temperature of 298 K is desired, the temperature of the plenum chamber is set to 298 K and that of the burner plate to 348 K. The oil is applied to the burner plate in order to maintain the plate at a constant temperature and to heat up the cooler unburned gas mixture as it flows through the plate, which compensates for the heat loss necessary to stabilize the flame.

Flow rates for liquid compounds are measured using a Bronkhorst mini-CORI-FLOW Mass Flow Controller connected to a Controlled Evaporator Mixer (CEM). The CEM is connected to the plenum chamber using a duct which is wrapped with heating cable in order to keep the compound in the gaseous state as it enters the chamber. Temperature controllers indicate the temperature of the experimental setup at two locations: one below the chamber after the Mass Flow Controllers and one inside the chamber below the burner plate.

Flow rates for gaseous compounds are measured using Bronkhorst High-Tech Mass Flow Controllers (MFCs). At most conditions, the MFC used for oxygen has a maximum flow of 3 nL/min (normal litres per minute) while that used for nitrogen has a maximum flow of 15 nL/min. When the adiabatic flame burning velocity of a gas mixture is above a certain limit, specific to each different temperature tested, two MFCs are used to allow a sufficient flow of oxygen through the burner plate. Bronkhorst conversion coefficient factors are used to convert an MFC designed for nitrogen (with a maximum flow of 3 nL/min) for use with a flow of oxygen.

Adiabatic flame burning velocities in a flat-flame burner are measured using the radial temperature distribution on the surface of the burner plate. If the sum of the heat loss and heat gain is larger than zero, the gas velocity is lower than the adiabatic flame burning velocity. Thus, the centre of the burner plate is hotter than the circumference and the flame is stabilized under sub-adiabatic conditions. Conversely, if the sum of the heat loss and heat gain is lower than zero, the flame is stabilized under super-adiabatic conditions and the centre of the plate is cooler than the circumference. When the flow rate of the gas mixture is changed, an appropriate value of the gas velocity is found that will cancel out the net heat flux. Therefore, the heat loss from the flame to the burner plate is compensated by the heat flux from the burner plate to the unburned gas mixture. The radial temperature distribution in the burner plate is uniform and equal to the temperature of the heating jacket. At this point, the flame is adiabatic, the sum of the heat loss and heat gain from the flame is zero and the adiabatic burning velocity is measured.

Uncertainties in determination of the flame burning velocity are due to errors in:

- Mass flow measurements, with a maximum error of 0.5% per MFC, as given by the manufacturer, Bronkhorst, and a maximum global error in laminar flame velocity of 1.5%.
- The thermocouples, with an error of 0.2 cm s^{-1} in the laminar flame velocity. This error was measured by changing the gas flow rate by small amounts that did not affect the uniform temperature profile of the flame on the burner plate noticeably.
- Flame distortions, for example edge effects, with an error of 0.2 cm s^{-1} in the laminar flame velocity in the range of equivalence ratios studied. This error was measured similar to the error in the thermocouples.

Errors of around 1% were present when determining the equivalence ratio, Φ , due to mass flow effects, as given by Bronkhorst. The temperature of the unburned gas mixture before and after it flowed through the plenum chamber was measured with a thermocouple. An error of 2 K was estimated. Errors in gas and liquid fuel purity were not significant as high purity compounds were used.

3. Results

3.1. Flat-Flame Velocities of Diethyl Ether

The apparatus has previously been validated with methane, which is described in detail by Dirrenberger *et al.* [28]. Diethyl ether was studied in air at a pressure of 1 atm, initial temperatures between 298 and 398 K and equivalence ratios from 0.55 to 1.60, Fig. 2. Error bars have been added to these data points. The adiabatic burning velocity increases sequentially with an increase in initial temperature. The maximum flame burning velocity is recorded at $\Phi = 1.15$ at 398 and 378 K and at $\Phi = 1.10$ at 388, 358, 328 and 298 K. It appears that the maximum burning velocities are shifting to the higher equivalence ratio of 1.15 at higher temperatures, but this is not the case for a temperature of 388 K. This may be due to uncertainties in the flame burning velocity. The adiabatic burning velocity measurements obtained are shown in Table 1, below.

3.2. Comparison with Literature Data

In 1959, Gibbs and Calcote [23] measured the burning velocities of 77 different compounds in order to show the effect of molecular structure on these velocities. Burning velocities of diethyl ether were determined at 298 K and atmospheric pressure, Fig. 3. They found the maximum burning velocity to be at $\Phi = 1.0$ with a value of 48.0 cm s^{-1} , which is in contrast with the results obtained in *this study* which show a maximum burning velocity of 41.81 cm s^{-1} at $\Phi = 1.10$. This is due to the fact that Gibbs and Calcote did not take into account stretch correction of their flames, as most modern studies do. In addition, Davis and Law [29] have reported that the flames studied by Gibbs and Calcote [23] were non-adiabatic and non-equidiffusive, which are factors that have a great effect on the laminar flame burning velocity of the fuel. Therefore, their results are consistently different from those obtained with more modern techniques.

Di *et al.* [24] measured laminar burning velocities of diethyl ether at different temperatures, pressures and equivalence ratios using spherically propagating flames in a combustion vessel. The maximum burning velocity at atmospheric pressure and at all temperatures is recorded at $\Phi = 1.10$, which is comparable with measurements from *this study* and results simulated using the mechanism of Yasunaga *et al.* [20], Fig. 4. However, the results of Di *et al.* [24] are quite scattered and deviate significantly from the results obtained in *this study* and the simulated results, especially at high initial temperatures. Under lean conditions, velocities measured by Di *et al.* [24] are faster than those obtained in *this study* and the simulated results, whereas under rich conditions, velocities are slower than those in *this*

study and the simulated results. Variations in these data sets may be due to the stretch correction necessary to acquire laminar flame velocities in combustion vessels with spherical flames. In the case of spherically propagating flames, two conditions occur which must be accounted for [30]: the rise in pressure in the combustion vessel must be determined correctly and the equation used to measure flame velocities must be valid throughout this pressure rise.

3.3. Comparison with Model

Adiabatic flame velocity measurements obtained in *this study* were compared to simulated results obtained using the mechanism of Yasunaga *et al.* [20], updated with a new C₀–C₃ sub-mechanism in construction at the Combustion Chemistry Centre in NUI Galway, Fig. 5 and 6. The C₀–C₃ sub-mechanism is based on our previous work [31–33], with the most recent updates described in [34, 35]. The experimental results are well-predicted by the model and the overall trend in flame velocities is reproduced. Deviations between the simulated and the experimental results are within the limits of experimental errors. All simulations were performed using Chemkin-PRO [36].

Sensitivity analyses were performed using the sensitivity function available in Chemkin-PRO [31] and show the effect of a reaction on the flow rate, which corresponds to the burning velocity. These analyses were carried out at an equivalence ratio of 1.10 and an initial temperature of 298 K, Fig. 7, and at an equivalence ratio of 1.15 and an initial temperature of 398 K, Fig. 8. Negative reactions inhibit reactivity and positive reactions promote reactivity. The chain branching reaction between hydrogen atom and molecular oxygen to produce oxygen atom and hydroxyl radical is omitted for clarity and has a value of +0.406 at 298 K and $\Phi = 1.10$ and a value of 0.411 at 398 K and $\Phi = 1.15$. Of the 20 most important reactions, those with smaller molecules, such as methyl, ethyl and carbon monoxide, have the largest effect on the flame burning velocities of diethyl ether. It is important to note that only one reaction involving diethyl ether has a significant effect on flame burning velocities, that between diethyl ether and hydrogen atom to form hydrogen molecule and the secondary radical, C₂H₅OC₂H₄-s. The reaction between hydrogen atom and hydroxyl radical to produce water is the most inhibiting reaction at both conditions because it is in direct competition with the chain branching reaction to produce oxygen atom and hydroxyl radical.

4. Conclusions

Adiabatic flame burning velocities of diethyl ether in air have been studied on a flat-flame burner at atmospheric pressure and six temperatures between 298 and 398 K. Equivalence ratios for these experiments ranged from 0.55 to 1.60. It was found that the adiabatic burning velocities increased with respect to temperature. The maximum flame velocity was reported to be at $\Phi = 1.10$ at 298, 328, 358 and 388 K and at $\Phi = 1.15$ at 378 and 398 K. It was suggested that the maximum velocities were shifting to $\Phi = 1.15$ at higher temperatures, but this was not the case at 388 K, due to uncertainties. These results were compared with experimental results from the literature [23, 24] and were found to deviate significantly. The results of Gibbs and Calcote [23] were non-adiabatic and non-equidiffusive and stretching of the flame was not accounted for. The results of Di *et al.* [24] differed from the findings of *this study*, possibly due to the fact that stretch correction was necessary to obtain the laminar burning velocities. A recently constructed chemical kinetic mechanism for diethyl ether oxidation and pyrolysis [20] was compared to experimental results and excellent agreement was achieved. It was found that the flame burning velocities reported in *this study* compared well to the simulated results. Sensitivity analyses were performed and small species, such as methyl, ethyl and carbon monoxide, were reported to have the greatest effect on the laminar burning velocities of diethyl ether.

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Figure 1.
Schematic of flat-flame adiabatic burner [28].

Figure 2.
Adiabatic burning velocities of diethyl ether with error bars.

Figure 3.
Comparison of diethyl ether data obtained in *this study* with data obtained by Gibbs and Calcote [23] at 298 K.

Figure 4.

Comparison of diethyl ether data obtained in *this study* at 328 and 358 K with data obtained by Di *et al.* [24] at 323 and 363 K. Results simulated using the mechanism of Yasunaga *et al.* [20]. Symbols: experiment; lines: simulation.

Figure 5. Comparison of diethyl ether data obtained in *this study* with results simulated using the mechanism of Yasunaga *et al.* [20] at 298, 358 and 388 K. Symbols: experiment; lines: simulation.

Figure 6.

Comparison of diethyl ether data obtained in *this study* with results simulated using the mechanism of Yasunaga et al. [20] at 328, 378 and 398 K. Symbols: experiment; lines: simulation.

Figure 7.
Sensitivity coefficients showing the 20 most important reactions in diethyl ether flame burning velocities at 298 K and $\Phi = 1.10$.

Figure 8.
Sensitivity coefficients showing the 20 most important reactions in diethyl ether flame burning velocities at 398 K and $\Phi = 1.15$.

Table 1

Adiabatic burning velocities (in cm s^{-1}) of diethyl ether obtained in the flat-flame burner. Maximum burning velocities at each equivalence ratio are in bold.

Φ ($\pm 1\%$)	298 K ($\pm 2\text{K}$)	328 K ($\pm 2\text{K}$)	358 K ($\pm 2\text{K}$)	378 K ($\pm 2\text{K}$)	388 K ($\pm 2\text{K}$)	398 K ($\pm 2\text{K}$)
0.55	9.04 (± 0.54)	11.32 (± 0.57)	13.24 (± 0.60)	16.96 (± 0.65)	18.12 (± 0.67)	18.61 (± 0.68)
0.60	13.54 (± 0.60)	16.29 (± 0.64)	19.48 (± 0.69)	22.79 (± 0.74)	24.13 (± 0.76)	25.22 (± 0.78)
0.65	17.72 (± 0.67)	21.15 (± 0.72)	24.86 (± 0.77)	28.34 (± 0.83)	30.08 (± 0.85)	31.45 (± 0.87)
0.70	21.97 (± 0.73)	25.88 (± 0.79)	29.82 (± 0.85)	33.70 (± 0.91)	35.49 (± 0.93)	37.26 (± 0.96)
0.75	25.68 (± 0.79)	30.39 (± 0.86)	34.85 (± 0.92)	39.02 (± 0.99)	41.01 (± 1.02)	42.87 (± 1.04)
0.80	29.03 (± 0.84)	34.81 (± 0.92)	39.12 (± 0.99)	44.01 (± 1.06)	46.12 (± 1.09)	48.24 (± 1.12)
0.85	32.74 (± 0.89)	38.20 (± 0.97)	43.55 (± 1.05)	48.55 (± 1.13)	51.18 (± 1.17)	53.01 (± 1.20)
0.90	35.70 (± 0.94)	41.84 (± 1.03)	48.11 (± 1.12)	52.22 (± 1.18)	55.14 (± 1.23)	57.22 (± 1.26)
0.95	38.25 (± 0.97)	44.31 (± 1.06)	50.97 (± 1.16)	55.64 (± 1.23)	58.39 (± 1.28)	60.47 (± 1.31)
1.00	40.13 (± 1.00)	46.43 (± 1.10)	53.25 (± 1.20)	58.13 (± 1.27)	60.98 (± 1.31)	62.89 (± 1.34)
1.05	41.28 (± 1.02)	47.79 (± 1.12)	54.64 (± 1.22)	59.50 (± 1.29)	62.25 (± 1.33)	64.22 (± 1.36)
1.10	41.81 (± 1.03)	48.54 (± 1.13)	55.51 (± 1.23)	60.30 (± 1.30)	63.10 (± 1.35)	65.26 (± 1.38)
1.15	41.49 (± 1.02)	47.87 (± 1.12)	55.44 (± 1.23)	60.32 (± 1.30)	62.61 (± 1.34)	65.49 (± 1.38)
1.20	40.24 (± 1.00)	46.98 (± 1.10)	54.25 (± 1.21)	59.08 (± 1.29)	61.37 (± 1.32)	64.16 (± 1.36)
1.25	38.70 (± 0.98)	45.15 (± 1.08)	52.28 (± 1.18)	56.85 (± 1.25)	59.10 (± 1.29)	61.78 (± 1.33)
1.30	36.73 (± 0.95)	42.46 (± 1.04)	49.32 (± 1.14)	53.74 (± 1.21)	55.94 (± 1.24)	57.99 (± 1.27)
1.35	33.03 (± 0.90)	39.50 (± 0.99)	45.51 (± 1.08)	49.52 (± 1.14)	51.64 (± 1.17)	54.01 (± 1.21)
1.40	29.81 (± 0.85)	35.23 (± 0.93)	41.23 (± 1.02)	45.26 (± 1.08)	47.09 (± 1.11)	49.56 (± 1.14)
1.45	25.95 (± 0.79)	30.88 (± 0.86)	36.34 (± 0.95)	39.89 (± 1.00)	42.56 (± 1.04)	43.96 (± 1.06)
1.50	22.12 (± 0.73)	26.64 (± 0.80)	31.45 (± 0.87)	34.76 (± 0.92)	36.96 (± 0.95)	39.08 (± 0.99)
1.55	18.56 (± 0.68)	22.44 (± 0.74)	26.44 (± 0.80)	29.89 (± 0.85)	30.96 (± 0.86)	32.81 (± 0.89)
1.60	15.46 (± 0.63)	18.28 (± 0.67)	21.97 (± 0.73)	24.41 (± 0.77)	25.91 (± 0.79)	26.26 (± 0.79)