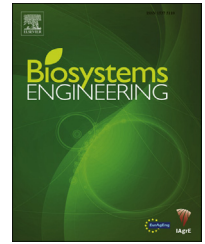


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Research Paper

Experimental study of the ignition delay of diesel/biodiesel blends using a shock tube



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Ignition delays of a pure biodiesel, which is produced from palm oil, as well as its blends with petroleum diesel were experimentally quantified using a preheated shock tube. The emission of OH^{*} radical signals, which was observed by a photomultiplier via a monochromator, was used to identify the time for onset of ignition. Experiments were performed behind the reflected shock waves at a pressure of 0.12 MPa, equivalence ratios of 0.5, 1.0 and 1.5, and a range of temperatures from 1174 to 1685 K. Fuel blends B0, B20, B40, B60, B80 and B100 (corresponding to 0, 20, 40, 60, 80 and 100 vol% of biodiesel with petroleum diesel, respectively) were tested. The results show that ignition delay variations of blends versus temperature were similar to those of pure diesel fuel. It was consistently found that for all fuel blends, ignition delay increases with an increase in equivalence ratio. An equivalence ratio exponent of 0.73 in Arrhenius correlation was observed. At a constant equivalence ratio, the effect of biodiesel fraction on chemical ignition delay of the fuel blends was not significant. The overall activation energy of diesel/biodiesel mixtures in this study is 161,937.5 J mol⁻¹.

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1. Introduction

With more world-wide energy consumption and increased shortages of fossil-based fuels, much research has focused on energy saving and reducing environmental pollution. Biodiesel is a clean burning alternative fuel produced from renewable resources such as vegetable oils and fats (Shao, He, Sun, & Jiang, 2009). It is becoming one of the most promising alternative fuels for global energy demands (Bo & Rosnah, 2014). Blends of biodiesel with petroleum diesel have been tested in diesel engines (Hifjur & Sweeti, 2014) and showed reductions in tailpipe emissions such as soot, carbon dioxide (CO₂), carbon monoxide (CO), unburned hydrocarbons (HC)

and NO_x without significant changes having to be made to the engine design.

Biodiesel is long-chain mono alkyl ester mixture which is converted from plant oils, recycled cooking greases and oils, or animal fats. It usually consists of fatty acid methyl esters (FAME) produced through a process called transesterification.

Ignition delay is the one of important parameters in combustion process of fuels, it directly impacts on the heat release rate and the timing of the onset of ignition in the thermodynamic cycle of an engine. It also indirectly affects engine performance, noise generation and pollutant formation. Measurements used shock tube facilities to obtain the chemical ignition delays of diesel and biodiesel are reviewed as follows.

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Nomenclature

Abbreviation

B0	Pure diesel
B20	Blend of 80% diesel with 20% biodiesel
B40	Blend of 60% diesel with 40% biodiesel
B60	Blend of 40% diesel with 60% biodiesel
B80	Blend of 20% diesel with 80% biodiesel
B100	Pure biodiesel
CEE	Cotton ethyl ester
CH*	Hydrocarbon radical
C/H/O	Fraction of carbon, hydrogen and oxygen in fuel
FAME	Fatty acid methyl esters
HC	Unburned hydrocarbons
H/C	Ratio of hydrogen and carbon
HPLC	High performance liquid chromatography
MLR	Multiple linear regression
OH*	Hydroxylation radical
PME	Palm methyl ester

Symbols

A	Coefficient determined from multiple linear regression method
E_a	Global activation energy, J mol ⁻¹
R	Universal gas constant, J mol ⁻¹ K ⁻¹
p	Pressure, MPa
T	Temperature, K
α	Pressure exponent
β	Temperature exponent
τ	Ignition delay, μ s
ϕ	Fuel-air equivalence ratio

Ignition delay of three large normal alkanes (n-decane, n-dodecane and n-hexadecane); methyl decanoate; and several diesel types (DF-2 with a range of cetane indices from 42 to 55) was studied using a shock tube (Haylett, Davidson, & Hanson, 2012). At high temperature, the measured ignition delay of lean mixtures was significantly longer than those of rich mixtures, and their measured ignition delay of rich mixtures was a little bit shorter than their model predictions. Ignition delay of methyl oleate and methyl linoleate (biodiesel surrogates) at very low vapour pressure was measured by Campbell, Davidson, Hanson, and Westbrook (2013) using an aerosol shock tube. It was reported to be similar over the experimental conditions. Differences between experimental and computed results were strongly related to existing errors and uncertainties in the thermochemistry of the large methyl ester species. Ignition delay of DF-2 diesel/21% oxygen/argon mixtures was measured by Haylett, Lappas, Davidson, and Hanson (2009) using an aerosol shock tube. Their measurements provide an accurate database for validation on the kinetic mechanisms of diesel fuel and surrogates. Lancheros et al. (2012) measured ignition delay of surrogate biodiesel fuels in a high-pressure shock tube. Their kinetic mechanism yields improved predictions of profiles measured earlier and it also agreed fairly well with the experimental data over the conditions in this study. Saleh (2011) carried out an

experimental study for ignition delay of cotton methyl ester, cotton ethyl ester (CEE) and CEE – diesel blends from neat cottonseed oil using the shock tube, he found that the minimum ignition delay was observed at an equivalence ratio of 1.05 for all the tested fuels.

Although the previous studies for ignition delay of diesel and biodiesel fuel are numerous, the data obtained are not sufficient to fully understand their combustion characteristics due to the diversity of biodiesel and complexity of their detailed kinetic mechanisms. Many kinetic mechanisms for biodiesel have been developed recently (Herbinet, Pitz, & Westbrook, 2008; Herbinet, Pitz, & Westbrook, 2010; Olchanski & Burcat, 2006; Wang et al., 2010), but these mechanisms still need to be validated by experimental data. Therefore, it is necessary to continue with experimental investigations in order to improve our understanding of the auto-ignition characteristics of biodiesel and its blends.

2. Experimental setup

The experiments were conducted behind the reflected shock wave in a preheated stainless-steel shock tube (School of Energy and Power Engineering, Xi'an Jiaotong University, <http://epe.xjtu.edu.cn/en/>). The shock tube and its equipped facilities were schematically shown in Fig. 1 (Thi, Zhang, Fu, Huang, & Zhang, 2014; Thi, Zhang, & Huang, 2014).

The shock tube has a 2 m long driver section and a 5.3 m long driven section with an internal diameter of 115 mm. Double polycarbonate diaphragms divided the driver and driven sections before each experiment. Thickness of the diaphragms was selected according to magnitude of the reflected pressure. A mixture of helium and nitrogen with different fractions was used as the driver gas to obtain a longer test time. The shock tube was evacuated to a pressure below 0.02 Pa before mixture of He and N₂ was introduced into the driver section and reactant mixture was added into the driven section. Four fast-response sidewall piezoelectric pressure transducers (PCB 113B26 – <http://www.pcb.com/Products.aspx?m=113B26>) are located at fixed intervals (300 mm) along the end part of driven section. Three time counters (FLUKE PM6690 – <http://www.ttid.co.uk/products-resale/fluke/fluke-counters-6690-spec.htm>) were used to record time intervals when the incident shock wave passed each transducer and the incident shock velocity was then correspondingly calculated.

The shock wave velocity at the end-wall was calculated by linear extrapolating the shock velocity profile to the end-wall, velocity uncertainty is approximately 0.2%. Typical attenuation rates of the incident shock ranged from 0.3 to 0.6 % m⁻¹. On the end-wall, a 5th pressure transducer (PCB 113B03 – <http://www.pcb.com/products.aspx?m=113B03>) was installed to measure the pressure behind the reflected shock wave, uncertainty in the pressure measurement was approximately 1%. Additionally, a photomultiplier (Hamamatsu CR131 <http://www.hamamatsu.com/eu/en/index.html>) and a quartz-glass window together with a 307.8 nm narrow band pass filter were mounted at the same position to capture OH* emission.

Temperature behind the reflected shock wave was calculated through the measured shock wave velocity by using the

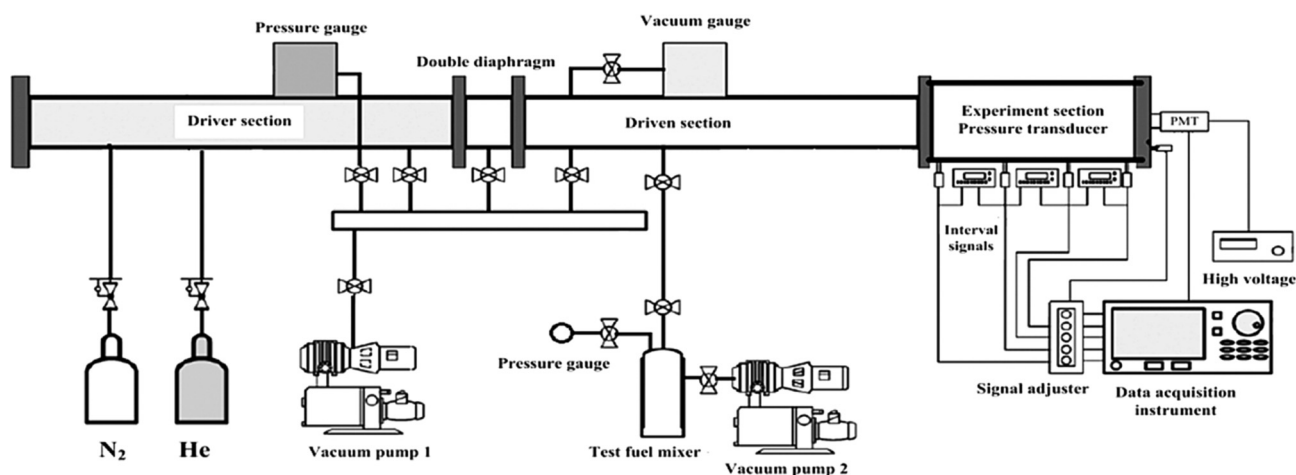


Fig. 1 – Schematic diagram of shock tube facility.

chemical equilibrium software GasEq (<http://www.c.morley.dsl.pipex.com/>), assuming frozen chemistry, with the thermodynamic data suggested by Burcat and Ruscic (2010). The uncertainty of the reflected shock temperature was less than 20 K, using a standard error analysis procedure (Zienkiewicz & Zhu, 1987).

The biodiesel used in this study was derived from residues of a process of refining cooking oil from palm oil. The production process was developed through a previous project in Vietnam (Nguyen, Vu, Do, Woo, & Jun, 2013). A commercial diesel fuel (diesel N°0) is used in this study. The biodiesel (palm methyl ester - PME) was produced from palm oil and methanol by a transesterification process in the presence of an alkali as a catalyst (NaOH or KOH). B0 (pure diesel), B20, B40, B60, B80 and B100 (pure biodiesel) were used in this study which correspond to 0, 20, 40, 60, 80 and 100% of biodiesel volume fractions in biodiesel-diesel mixtures, respectively. Properties of diesel and biodiesel fuels are given in Table 1.

Table 1 – Properties of the diesel and biodiesel fuels.

Fuels	Diesel	Biodiesel
H/C ratio (moles)	1.788	1.902
Low heat value [MJ kg^{-1}]	42.92	37.39
Molecular weight [g mol^{-1}]	191.8	295.31
Cetane number	49	66.9
Density (15 °C) [g ml^{-3}]	0.8369	0.8693
Viscosity (40 °C) [$\text{mm}^2 \text{s}^{-1}$]	3.14	4.1
Acid Number [mg KOH g^{-1}]	0.023	0.06
Flash Point [°C]	60	152
Cloud Point [°C]	3	18

Table 2 – Fractions of B0 and B100.

Fuel	C [%]	H [%]	O [%]	Others [%]
B0	86.93	12.96	0.07	0.04
B100	76.96	12.17	10.83	0.04

The C/H/O fractions of these diesel and biodiesel fuels were determined separately by a high performance liquid chromatography technique (HPLC). The experimental results measured the C/H/O fractions for B0 and B100 are shown in Table 2.

The fuel mixtures were prepared according to the partial pressure of each gas component in a 128 l stainless steel tank, which was also evacuated to pressure below 0.02 Pa before filling with the test mixtures. After mixing, the mixtures were settled over night to ensure perfect mixing. Because of the low

Table 3 – Compositions of the test mixtures.

Blends	Diesel (% mol)	Biodiesel (% mol)	Oxygen (% mol)	Nitrogen (% mol)
B0	1.03	0	20.79	78.18
B20	0.84	0.14	20.8	78.22
B40	0.64	0.29	20.82	78.25
B60	0.44	0.44	20.82	78.3
B80	0.22	0.6	20.84	78.34
B100	0	0.77	20.85	78.38

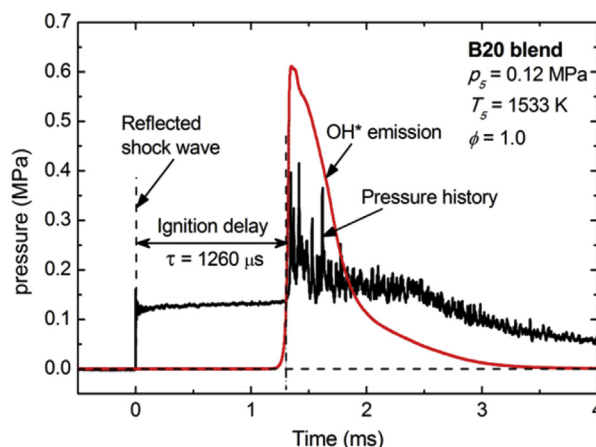


Fig. 2 – Definition of ignition delay.

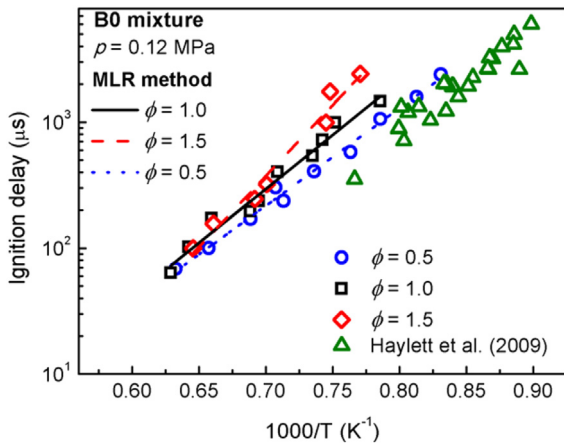


Fig. 3 – Ignition delay of diesel (B0) in air at pressure of 0.12 MPa, $\phi = 0.5, 1.0$ and 1.5; and the ignition delay data of Haylett et al. (2009) normalised to $p = 0.12$ MPa and $\phi = 0.5$ using p^{-1} and ϕ^{-1} .

saturated vapour pressures of diesel and biodiesel, both the shock tube driven section and the mixing tank were uniformly heated up to 393 K and maintained at that temperature by electric heating elements to ensure complete vaporisation of all liquid fuels. Detailed compositions of the stoichiometric test mixtures are given in Table 3.

Previous studies showed that the emission signal from either OH^* or CH^* radical is the simplest and the most reliable

method of diagnosing the ignition of combustible mixtures (Petersen et al., 2007; Rickard, Hall, & Petersen, 2005). Ignition delay is defined as the time interval between the arrival of the shock wave at the end-wall and the onset of the ignition, which was determined by extrapolating the steepest rise in the OH^* chemiluminescence signal to the zero baseline, as shown in Fig. 2.

3. Results and discussion

All experiments for six different fuel blends (B0, B20, B40, B60, B80 and B100) were conducted in the range of temperature from 1174 K to 1685 K, pressure of 0.12 MPa, and equivalence ratios of 0.5, 1.0 and 1.5. Nitrogen was used as the diluted gas, and in the all test mixtures, a constant volume ratio of 3.76 is maintained for oxygen and nitrogen which exhibits the typical proportion of these two gases at atmosphere. Data of the ignition delay under all test conditions are provided in Supplementary Material to this paper. Results of the ignition delay are shown in Figs. 2 and 3.

Ignition delay exhibits an Arrhenius-type behaviour with respect to temperature, indicating the typical Arrhenius dependence of ignition delay on temperature. The following empirical correlations were obtained using the multiple linear regression (MLR) method,

$$\tau = Ap^{\alpha}\phi^{\beta} \exp\left(\frac{E_a}{RT}\right) \quad (1)$$

Table 4 – Correlation parameters for diesel/biodiesel blends in Eq. (1).

Blends	$\phi = 0.5$		$\phi = 1.0$		$\phi = 1.5$	
	A	E_a (J mol ⁻¹)	A	E_a (J mol ⁻¹)	A	E_a (J mol ⁻¹)
B0	0.00084	148,000.6	0.0003	163,920.8	0.000005	215,354.7
B20	0.0002	160,757.6	0.0015	146385.6	0.000056	190,493.3
B40	0.00015	167,418.6	0.0013	144,900.3	0.0015	148,281
B60	0.00087	146,615.7	0.000028	191,079.1	0.000124	179,167.2
B80	0.00043	153,469.1	0.0015	142,992.4	0.000087	181,209
B100	0.00018	162,979.4	0.00011	175,012.5	0.0001	178,502

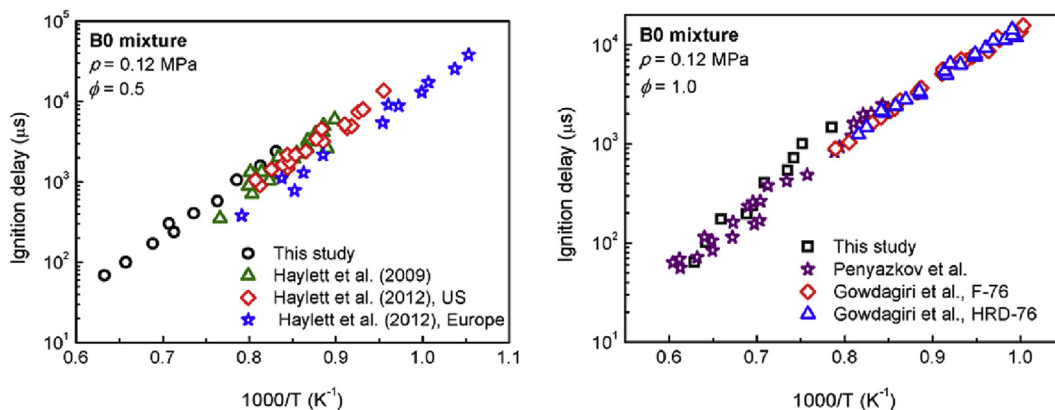


Fig. 4 – Ignition delay of diesel (B0) in air at pressure of 0.12 MPa, $\phi = 0.5$ and 1.0 from this study with diesel measurements normalised to $p = 0.12$ MPa from Haylett et al. (2009, 2012) (using $p^{-0.82}$), Penyazkov et al. (2009) (using p^{-1}) and Gowdagiri et al. (2014) (using p^{-1}).

The correlation parameters for each mixture condition are given in Table 4, where $R = 8.314 \text{ J mol}^{-1} \text{ K}^{-1}$ is universal gas constant; E_a is global activation energy for the combustion process in J mol^{-1} ; ϕ is fuel-air equivalence ratio; T and p are temperature (K) and pressure (MPa), respectively; α , β and A are constants determined from MLR method; and τ is ignition delay in microsecond. It should be noted that Eq. (1) is only applicable for empirically calculating the ignition delay under the test conditions used in this study. Under different test conditions, the Arrhenius dependence of ignition delay on temperature could be different.

Figure 3 gives the ignition delay of neat diesel in air at equivalence ratios of 0.5, 1.0 and 1.5 and pressure of 0.12 MPa.

The measured ignition delay of Haylett et al. (2009) (corresponding to temperature which is higher than 1100 K), normalised to pressure of 0.12 MPa and equivalence ratio of 0.5 using their exponents which were found to be -1 , is also presented for comparison. It is clear from Fig. 3 that the ignition delay of neat diesel in this study and of Haylett et al. (2009) exhibits clear Arrhenius dependences on temperature at equivalence ratios. A comparison of ignition delay data at an equivalence ratio of 0.5 in present study (blue colour in web version) with that from Haylett (green colour in web version) in Fig. 3 shows that the two data were almost consistent, the between-two data error is less than 20%. The error can be derived from the different experimental conditions and the

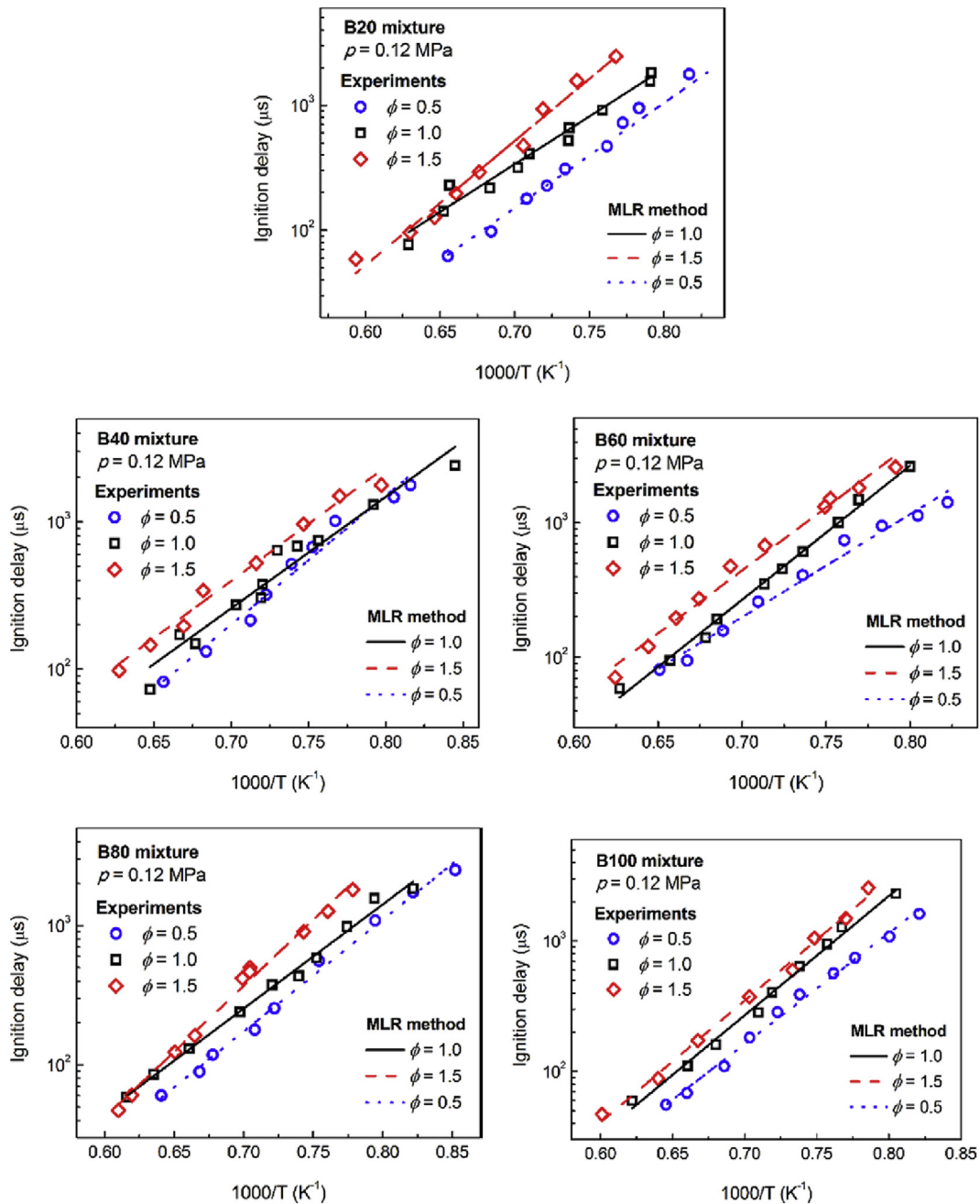


Fig. 5 – The experimental ignition delay of diesel/biodiesel blends. All lines represent fits to the corresponding data.

different dilute gases. The overall activation energy of diesel mixture in this study is $148,000.6 \text{ J mol}^{-1}$, meanwhile, it is equal to $150,741.2 \text{ J mol}^{-1}$ found in the work of Haylett. Obviously, these two overall activation energies are similar.

The ignition delays in this study at equivalence ratios of 0.5 and 1.0 are compared to the previous studies of diesel fuel ignition by Haylett et al. (2009, 2012) and Penyazkov, Sevrouk, Tangirala, and Joshi (2009) and Gowdagiri, Wang, and Oehlschlaeger (2014) in Fig. 4.

The results shown in Fig. 4 indicate that ignition delay found in this study is in close agreement with all previous studies, irrespective of fuel composition. The measurements of Haylett et al. (2009, 2012) were carried out for DF-2 diesel, the experiments of Penyazkov et al. (2009) were performed for DF-2 diesel/air mixtures, and Gowdagiri et al. (2014) studied the military diesel F-76 and renewable diesel derived from algal oils, HRD-76. The overall activation energies of these diesel/air mixtures were almost similar, indicating that the previous measurement methods and the shock tube facility used here are reliable and accurate.

Figure 5 gives the ignition delay of diesel/biodiesel blends (B20, B40, B60, B80 and B100) at equivalence ratios of 0.5, 1.0 and 1.5 and pressure of 0.12 MPa. It can be seen that the ignition delay of each mixture also exhibited a clear Arrhenius dependence versus temperature at all equivalence ratios. The slope of straight lines of ignition delay versus inverse temperature at different equivalence ratios represents the overall activation energy of the reactant mixture. The average overall activation energy of all blends is approximately $163,176 \text{ J mol}^{-1}$, as shown in Table 4. It is to be noted that the MLR method is only applied to each fuel/air mixture, therefore, the pressure and equivalence ratio parameters in Eq. (1) do not need to be taken, because their exponents (α , β) are constant and the product $p^\alpha \phi^\beta$ is integrated into coefficient A. This value is in nearly agreement with the parameters of E_a for methyl oleate and methyl linoleate reported in (Campbell et al., 2013) (Tables 1 and 2), the small difference in activation energy could be due to the different fuel types.

A comparison of B100 ignition delays carried out at pressure of 0.12 MPa, equivalence ratio of 0.5 is shown in Fig. 6. Although biodiesels have been produced from different feedstocks and studied at various conditions, the normalised ignition delays obtained are consistent.

For all diesel/biodiesel blends, ignition delay increases with the increase of equivalence ratio for all blends, however, the effect of equivalence ratio on ignition delay is fairly minimal. This is in agreement with previous findings in Campbell et al., (2013); Lancheros et al., (2012); and Saleh (2011).

Figure 7 shows that the effect of biodiesel fraction on ignition delay of the corresponding mixture is not significant. This demonstrates that the biodiesel addition insubstantially affects chemical delay, however, it could decrease the physical delay due to its physical properties, such as higher viscosity, higher specific gravity, lower compressibility, lower calorific value and lower volatility (Shahabuddin, Liaquat, Masjuki, Kalam, & Mofijur, 2013). Much research has been performed to determine ignition delay of diesel engine fuelled with biodiesel. Obtained results of the studies showed that biodiesel has a shorter ignition delay than that of diesel (Shahabuddin et al., 2013).

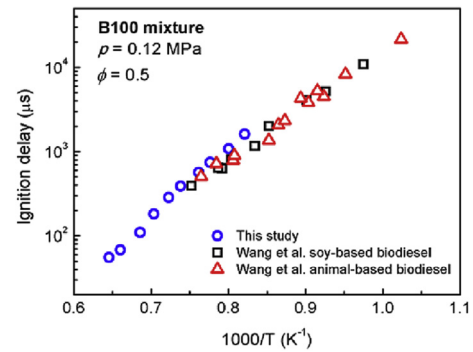


Fig. 6 – The ignition delay of B100 in this study and Wang, Gowdagiri, and Oehlschlaeger (2014). All data normalised to $p = 0.12 \text{ MPa}$ and $\phi = 0.5$ using $p^{-0.8}$ and $\phi^{-0.5}$ scaling.

Using experimental data recorded in this study together with MLR method to obtain an overall expression can calculate easily ignition delay of diesel/biodiesel/air mixtures. The fitted expression is presented as follows,

$$\tau = 3.47 \cdot 10^{-4} \cdot \phi^{0.73} \cdot \exp\left(\frac{161937.5}{RT}\right) \quad (2)$$

The positive exponent for the equivalence ratio demonstrates that the ignition delay is proportional to equivalence ratio as shown earlier in Figs. 2 and 3.

4. Conclusions

Shock tube ignition delay measurements for diesel/biodiesel/air mixtures at pressure of 0.12 MPa, equivalence ratios of 0.5, 1.0 and 1.5 and temperatures from 1174 K to 1685 K have been reported. The measurements supplement the accurate diesel/biodiesel blends ignition delay data set, which can be used to develop new combustion systems as well as to correct and

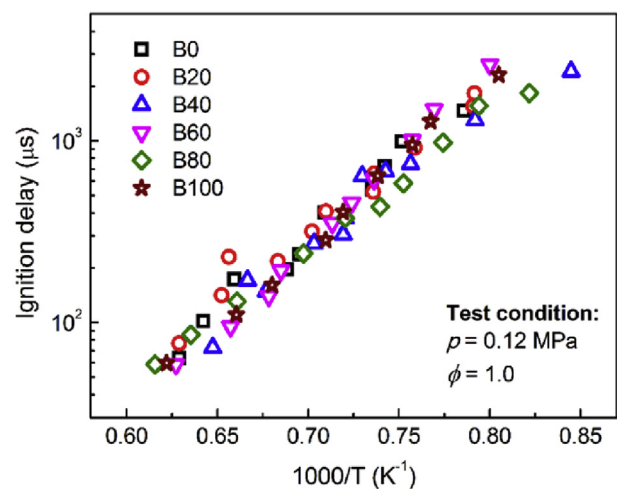


Fig. 7 – The experimental ignition delay of diesel/biodiesel blends in air at pressure of 0.12 MPa and equivalence ratio of 1.0.

validate biodiesel kinetic mechanisms. In addition, the data can also compare to ignition delays of the surrogate diesel fuel mixtures.

The measured results showed that ignition delay increases with increasing equivalence ratio for all blends, however, the effect of equivalence ratio on ignition delay is quite small since the equivalence ratio exponent is 0.73. The measured ignition delays are in good agreement with the previous results. The overall activation energy of diesel/biodiesel mixtures in this study was $161,937.5 \text{ J mol}^{-1}$.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.biosystemseng.2015.03.009>.

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