Iranian Journal of Chemical Engineering
Vol. 13, No. 1 (Winter 2016), IAChE

Research note

An Empirical Correlation to Predict the Ignition Delay Time for Some Hydrocarbon Fuels

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Received: June 2015 *Accepted: November 2015*

Abstract

Examination of the available ignition delay time data and correlations in the case of methane, butane, heptane, decane, kerosene, Jet-A and ethylene fuels, allowed the derivation and recommendation of standard equations for this property. In this study, a new accurate substance dependent equation for ignition delay time as a function of pressure, number of carbon atoms, mixture equivalence ratio, fuel mole fraction and temperature has been developed to estimate ignition delay time of some hydrocarbon fuels. With the presented model, ignition delay time has been calculated and compared with the data reported in literature. The accuracy of the obtained model has been compared to the mostly used predictive models and the comparison indicated that the proposed correlation provides more accurate results than other models used in the previous works.

Keywords: *Hydrocarbon Fuels, Ignition Delay Time, Shock Tube, Modeling, Correlation*

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1. Introduction

When a suitable fuel-oxidizer mixture is contained in a vessel, a chemical reaction will occur. If this reaction is an accelerating exothermic reaction, it will result in combustion accompanied by the formation of a visible flame. The time between the first contact of the reactants and the formation of a visible flame is named Ignition delay time [1-3]. Ignition delay of fuels is affected by many factors. The main parameters affecting on ignition delay are: temperature, pressure, fuel concentration, composition of fuel, oxidizer concentration, type of surface of reaction vessel, physical characteristics of vessel, the condition of mixture flow and method of fuel injection [1,3].

Although numerous ignition delay time studies were previously conducted, many hydrocarbon species relevant to practical fuels have not been extensively studied. Also, due to the large number of hydrocarbons that exist in practical fuels, it is clearly advantageous to develop a method that will reduce the number of experimental studies needed to determine the ignition delay characteristics of hydrocarbons. Establishing a method that enables ignition delay time measurements to be directly compared when obtained over different conditions would clearly be beneficial. A number of different methods have been employed for analyzing the ignition delay time measurements. Perhaps the most useful is the one that involves performing regression analysis on the experimental data, and using the resultant empirical regression coefficients to express the ignition delay time as a function of key parameters. The development of correlations facilitates the comparison of ignition delay time data among studies, and enables the ignition delay time sensitivity to a particular parameter to be explicitly stated [2,4-6]. Correlations also guide the experimentalists in the design of data sets by reducing the number of experimental conditions needed to fully examine the ignition behavior of a particular fuel [4,6].

Numerous ignition delay time studies have been conducted over a wide range of conditions and for a variety of fuels by using shock tubes. As an experimental device, shock tubes are widely used to investigate the chemical kinetic behavior of reactive mixtures. The shock tube is preferred as it is a simple and unique device that allows fuels to be exposed to different temperatures, pressures and residence times similar to engine conditions [7]. In most of the shock tube studies, the experimental results of ignition delay time have been correlated with particular empirical equations.

Meredith *et al*. [3] derived a correlation for more than 500 methane-oxygen ignition-delay measurements over a temperature range of 1250–2500 K:

$$\tau = 2.21\times10^{-14}\exp\left(45000/RT\right)[O_2]^{-1.05}[CH_4]^{0.33} \quad (1)$$

An experimental study was performed by Petersen *et al*. [8] to determine ignition delay times for CH_4/O_2/diluent mixtures. The temperatures and pressures used were 1040-1600 K and 35-260 atm, respectively. The CH_4/O_2/diluent mixtures had an equivalence ratio of 0.4, 3.0, or 6.0 with N_2, Ar or He as the bath gas. A comparison of the ignition

delay times for each mixture resulted in two expressions. The subsequent high temperature correlation was:

$$\tau_{ign} = 1.26\times10^{-14}[CH_4]^{-0.02}[O_2]^{-1.2}\exp(32700/RT) \quad (2)$$

And the corresponding low-temperature expression was:

$$\tau_{ign} = 4.99\times10^{-14}[CH_4]^{-0.38}[O_2]^{-1.31}\exp(18950/RT) \quad (3)$$

In another work, Petersen *et al*. [9] investigated various CH_4-O_2 mixtures in argon baths over 1250-2100 K and 8-85-atm. The resulting data were correlated using the following empirical expression:

$$\tau = 1.08\times10^{-14}[O_2]^{-1.12}[CH_4]^{0.36}\exp(47/RT) \quad (4)$$

Grillo *et al*. [10] have measured ignition delay times in homogeneous CH_4-O_2 and CH_4-O_2-N_2 mixtures diluted in argon. The mixtures were heated in a shock tube to the temperatures range of 1640-2150 K and pressures 1-6 atm. For both mixtures, the measured ignition delay times were correlated using the empirical expression:

$$\tau_i = 4.4\times10^{-15}\exp(52300/RT)[CH_4]^{0.33}[O_2]^{-1.03} \quad (5)$$

Horning *et al*. [2,5] measured ignition delay times of butane fuel over the temperature range of 1250-1750 K, pressure range of 1-6 atm and mixture compositions of 2-20% oxygen with an equivalence ratio of 0.5 to 2. A Regression analysis of measurements yielded the following correlations:

$$\tau = 7.63\times10^{-8}[n-C_4H_{10}]^{0.99}[O_2]^{-1.61}e^{42850/RT} \quad (6)$$

$$\tau = 3.57\times10^{-5}P^{-0.64}X_{O_2}^{-0.62}\Phi^{1}e^{41000/RT} \quad (7)$$

In addition, they measured heptane ignition delay times at the same condition [2, 5]. The data were correlated as follow:

$$\tau = 4.54\times10^{-8}[n-C_7H_{16}]^{0.95}[O_2]^{-1.58}e^{45000/RT} \quad (8)$$

$$\tau = 6.67\times10^{-6}P^{-0.61}X_{O_2}^{-0.68}\Phi^{0.96}e^{44600/RT} \quad (9)$$

Davidson *et al*. [4,5] found in their laboratory that for n-heptane, ignition delay times could be correlated well in the form of equation (9). Under these conditions, Burcat *et al*. [3,4,11] developed a correlation over the temperature range of 1150–1410 K, pressures of approximately 6–8 atm, and equivalence ratios of 0.5 and 1.0 [3,4,11].

$$\tau = 3.2\times10^{-12}\exp(35300/RT)[C_7H_{16}]^{0.2}[O_2]^{-1.1}[Ar]^{0.6} \quad (10)$$

Meredith *et al*. [3] reported the ignition delay time correlation for heptane as follows:

$$\tau = 6.76\times10^{-15}\exp(40160/RT)[C_7H_{16}]^{0.4}[O_2]^{-1.2} \quad (11)$$

The ignition delay of n-decane and oxygen was investigated by Olchanski *et al*. [12] for a series of mixtures ranging from 0.49 to 1.5% decane and 4.16 to 23.25% O_2 diluted in argon in a heated shock-tube. The temperature and pressure ranges were 1239–1616 K and 1.82–10 atm, respectively. An overall ignition delay equation was deduced for 144 experiments:

$$\tau = 10^{-12}\exp(34240/RT)[C_{10}H_{22}]^{0.6}[O_2]^{-1.305}[Ar]^{0.08} \quad (12)$$

Liang *et al*. [13-17] measured ignition delay times of China No. 3 aviation kerosene using a heated shock tube. Experimental

conditions covered a temperature range of 820–1500 K, at pressures of 5.5, 11 and 22 atm, equivalence ratios of 0.5, 1 and 1.5, and oxygen concentration of 20%. The correlations were obtained in the following form:

$$\tau = 3.2\times10^{-11}\,[Kerosene]^{0.22}\,[O_2]^{-1.09}\,\exp(69941/RT) \quad (13)$$

$$\tau = 4.72\times10^{-7}\,P^{-0.88}\,\Phi^{0.23}\exp(62092/RT) \quad (14)$$

Zhukov *et al*. [18-20] measured ignition delay times for mixtures of Jet-A with air at pressures of 10 and 20 atm. The measurements were performed for the lean, stoichiometric and rich mixtures (Φ=0.5;1;2) behind the reflected shock wave in the temperature range of 1040–1380 K. The experimental data was summarized in a single expression:

$$\tau_{ign} = 1.31\times10^{-3}.(P)^{-0.67}.\Phi^{-0.6}.\exp(30.4/RT) \quad (15)$$

Meredith *et al*. [3] tested three ethylene/oxygen/argon mixtures with equivalence ratios of 0.5, 0.75, and 1.0 at reaction pressures of 5–8 atm and temperatures ranging from 1125 to 1410 K. A least-square fit of the combined data was the follow expression:

$$\tau = 2.82\times10^{-17}\exp(35000/RT)[O_2]^{-1.2} \quad (16)$$

Baker and Skinner [21,22] conducted ethylene studies in argon over a wide range of equivalence ratios (Φ=0.13,0.5,1 and 2) at pressures of 3 and 12 atm covering a temperature range of 1050–1550 K. They obtained an overall correlation equation:

$$\tau = 10^{-19}[C_2H_4]^{0.3}[O_2]^{-1.1}[Ar]^{0.4}\,e^{34200/RT} \quad (17)$$

Hidaka *et al*. [21,23] studied ignition of ethylene at varied pressures of 1–5 atm and the temperature range of 1400–2300 K. They derived an experimental correlation as given below:

$$\log\tau_{CH}\,[O_2] = -11.45 + \frac{27.5\times10^3}{4.58T} \quad (18)$$

Petersen *et al*. [21,24] conducted ethylene experiments at pressures of 1–3 atm, argon diluents of 98–96% (Φ=0.5 and 1.0) and a temperature range of 1250–1700 K. They obtained the correlation:

$$\tau = 3.3\times10^{-7}[C_2H_4]^{0.19}[O_2]^{-0.95}[Ar]^{0.04}\exp(26600/RT) \quad (19)$$

In another work, Saxena *et al*. [21] investigated ethylene combustion at a temperature range of 1000–1650 K, at pressures of 2, 10 and 18 atm, and equivalence ratios of 3 and 1. The correlation obtained based on the data, is given as shown below:

$$\tau = 10^{-29.5405}T^{6.7}e^{23840.1/T}[C_2H_4]^{0.07}[O_2]^{-0.92}[Ar]^{0.41} \quad (20)$$

Different correlations forms have been previously employed for similar fuels. This subject makes it difficult to directly compare the results from different studies. The goal of this work is to obtain an empirical ignition delay time correlation for some hydrocarbon fuels which have individual models in literature.

2. The proposed ignition delay time correlation

This work tried to find an ignition delay time correlation with high accuracy compared to other models, which were mentioned above.

The available data in sources were used to propose a general model for some hydrocarbon fuels (methane, butane, heptane, decane, kerosene, Jet-A, and ethylene). After regression analysis on the available experimental data (75% of data bank), a new equation was suggested as follows:

$$\tau = \left[b_1 \times P^{b_2} \times C^{b_3} \times \phi^{b_4} \times x_{fuel}^{b_5} \times \exp\left(\frac{b6 + (b7 \times T)}{(b8 \times T) + b9}\right) \right] \cdot \left(\frac{b10 \times R \times T}{P}\right) \quad (21)$$

where b_1 is simply a scaling constant, pressure (P) is in atm, C is the number of carbon atoms in the molecule, Φ is the mixture equivalence ratio, X_{fuel} is the fuel mole fraction, T is temperature in Kelvin, R is the universal gas constant (1.987), and b_2 to b_{10} are the empirically determined regression coefficients (which are individual for every fuel) and have been presented in Table 1. b_2 to b_{10} are tuned coefficients that have been determined by using least square curve fitting method and Marquardt-Levenberg algorithm which minimize the sum of the squared differences between the values of the observed and predicted values of the dependent variables. Sigma Plot software (version 11) was used to find coefficients.

Table 1
Tuned coefficients of new proposed model.

Fuel	Coefficients									
	b_1	b_2	b_3	b_4	b_5	b_6	b_7	b_8	b_9	b_{10}
Methane	1.7519E-7	-0.7376	-0.2400	0.6083	-0.4826	54094.5897	-19.5472	-0.4459	1679.3432	-0.0266
Butane	0.0229	-0.5822	4.0321	1.9969	-0.7393	10911.7813	-4.5336	2.6952	-2733.9357	-0.0066
Heptane	0.0007	-0.3978	0.9230	0.8513	-0.3434	30731.1524	5.6314	2.6702	-348.2348	0.0299
Decane	1.5559E-5	-0.5420	-0.1547	0.8000	-0.4802	37670.3596	-23.7766	-1.1938	1934.3950	-0.0639
Kerosene	2.2980E-5	-0.2292	-0.3421	0.2376	-0.5377	45532.1242	-13.6846	0.1371	1577.4204	-0.7635
Jet-A	6.0713E-7	-0.6198	-1.6776	0.9037	-1.5976	45155.9612	-25.8386	-0.6299	1338.2044	-0.1001
Ethylene	1.1467E-7	-0.2018	-7.5454	0.7486	-1.1736	45484.8801	-27.1435	-0.7170	1310.6492	-0.0524

3. Result and discussion

The experimental ignition delay time data for mentioned fuels were collected from different investigations and have been summarized in Table 2. The proposed model resulted from correlating 75% of these data.

Table 2
Ignition delay time data for hydrocarbon fuels in shock tube.

Fuel	Data points	T (K)	P (atm)	Equivalence ratio (Φ)	τ_{ign} (μs)	Reference
Methane	-	1196-1722	2.54-477	0.5	12-445	[25]
	-	1041-1607	12-263.6	0.4-6	55-875	[8]
	62	1323-2096	8-86.8	0.5-4	6-908	[9]
Butane	10	1645-2020	1.75-5.76	1	40-670	[10]
	39	1352-1734	1.03-3.81	0.5-2	83-475	[2]
Heptane	31	1223-1427	4.1-7.78	0.5-1	88.5-844	[3]
	60	1329-1676	1.14-5.71	0.5-2	86-488	[2]

	21	1021.6-1588.4	14.78-17.05	0.5	133-5630	[7]
	30	1237-1616	1.8-10	1	21-792	[12]
Decane	25	792-1321	10-102	1	39-337	[26]
	6	1397-1516	1.22-1.26	1	124-480	[2]
Kerosene	80	822.3-1501.3	4.9-27.6	0.5-1	20.2-4919.3	[13]
	40	1043-1378	9.3-29.4	0.5-2	15-416	[20]
Jet-A	40	1043-1378	9.3-29.4	0.49-2.02	15-416	[27]
	44	874-1229	17.3-50.9	0.5-1	28-3109	[14]
	-	1125-1414	4.83-7.89	0.5-1	0.415-290	[3]
Ethylene	-	1073-2211	1.3-4.8	1	4.4-797.4	[28]
	121	1000-1828	1.9-20.2	1-3	12-4404	[21]
	54	1253-1572	1.1-3.98	1	66-248	[2]

To compare the accuracy of the proposed empirical correlation with available models, the average absolute relative deviation percentage (AARD%), average relative deviation (ARD%), and R^2 value were calculated. The mathematical definition of the parameters including AARD%, ARD%, and R^2 values are given as shown below:

$$AARD\% = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{\tau_{i,\exp}-\tau_{i,calc}}{\tau_{i,\exp}}\right|\times 100 \quad (22)$$

$$ARD\% = \frac{1}{N}\sum_{i=1}^{N}\frac{\tau_{i,\exp}-\tau_{i,calc}}{\tau_{i,\exp}}\times 100 \quad (23)$$

$$R^2 = 1-\frac{\sum_{i=1}^{N}\left(\tau_{i,\exp}-\tau_{i,calc}\right)^2}{\sum_{i=1}^{N}\left(\tau_{i,\exp}-\bar{\tau}\right)^2} \quad (24)$$

where $\tau_{i,exp}$ is the experimental ignition delay time, $\tau_{i,calc}$ is the estimated ignition delay time and $\bar{\tau}$ is the average value of the experimental ignition delay time. In Tables 3-9, the AARD%, ARD%, and R^2 values of ignition delay time for models have been presented. These statistical parameters were calculated from the experimental data given in Table 2.

Table 3

Statistical parameters of this study compared with other models for methane/O_2 mixtures.

		Ref. [9]	**Ref. [10]**	**Total data**
AARD%	Eq. 1	89.658	21.036	80.127
	Eq. 2	4235.539	2768.322	4031.759
	Eq. 3	574401.592	327279.244	540079.043
	Eq. 4	138.661	9.748	120.756
	Eq. 5	91.231	31.457	82.929
	This study	24.383	27.584	24.828
ARD%	Eq. 1	-53.993	11.323	-44.921
	Eq. 2	-4235.349	-2768.322	-4031.595
	Eq. 3	-574401.592	-327279.244	-540079.043
	Eq. 4	-129.317	0.133	-111.338
	Eq. 5	-84.046	-30.928	-76.668

	This study	-1.837	-6.929	-2.544
	Eq. 1	0.473	0.786	0.521
	Eq. 2	-1368.57	-470.943	-1231.29
	Eq. 3	-2762121	-3423364	-23920465
R^2	Eq. 4	-0.441	0.973	-0.224
	Eq. 5	-0.082	0.956	0.076
	This study	0.951	0.701	0.913

Table 4

Statistical parameters of this study compared with other models for butane/O_2 mixtures.

		ref. [2]	**Total data**
	Eq. 6	10.687	10.687
AARD%	Eq. 7	11.327	11.327
	This study	5.885	5.885
	Eq. 6	8.617	8.617
ARD%	Eq. 7	-1.711	-1.711
	This study	-0.257	-0.257
	Eq. 6	0.870	0.870
R^2	Eq. 7	0.936	0.936
	This study	0.977	0.977

Table 5

Statistical parameters of this study compared with other models for heptane/O_2 mixtures.

		Ref. [2]	**Ref. [3]**	**Ref. [7]**	**Total data**
	Eq. 8	12.789	21.973	73.378	26.691
	Eq. 9	8.081	23.654	84.329	26.688
AARD%	Eq. 10	26.275	50.518	46.693	36.813
	Eq. 11	27.129	18.722	32.656	25.838
	This study	17.914	20.614	14.892	18.095
	Eq. 8	-9.328	-4.336	-33.218	-12.426
	Eq. 9	-1.442	-5.659	-44.828	-10.744
ARD%	Eq. 10	13.087	-49.814	-27.613	-11.954
	Eq. 11	-21.960	-12.034	19.171	-11.500
	This study	-9.913	-13.955	11.273	-7.059
	Eq. 8	0.991	0.818	-1.088	-0.859
	Eq. 9	0.995	0.773	-1.999	-1.668
R^2	Eq. 10	0.923	0.598	0.372	0.425
	Eq. 11	0.968	0.868	0.925	0.927
	This study	0.985	0.964	0.991	0.9900

Table 6
Statistical parameters of this study compared with other models for decane/O_2 mixtures.

		Ref. [2]	Ref. [12]	Ref. [26]	Total data
AARD%	Eq. 12	56.905	6.950	5269.833	2117.553
	This study	16.583	33.920	30.418	30.628
ARD%	Eq. 12	-56.905	0.243	-5269.833	-2114.022
	This study	-16.583	2.457	-18.198	-7.882
R^2	Eq. 12	0.045	0.983	-86821.9	-9528.33
	This study	0.922	0.715	0.722	0.729

Table 7
Statistical parameters of this study compared with other models for kerosene/O_2 mixtures.

		Ref. [13]	Total data
AARD%	Eq. 13	26.350	26.350
	Eq. 14	38.894	38.894
	This study	30.405	30.405
ARD%	Eq. 13	2.748	2.748
	Eq. 14	34.632	34.632
	This study	-9.793	-9.793
R^2	Eq. 13	0.601	0.601
	Eq. 14	0.544	0.544
	This study	0.849	0.849

Table 8
Statistical parameters of this study compared with other models for Jet-A/O_2 mixtures.

		Ref. [14]	Ref. [20]	Ref. [27]	Total data
AARD%	Eq. 15	32.592	14.017	14.013	20.607
	This study	11.357	20.930	21.005	17.558
ARD%	Eq. 15	12.811	-1.466	-1.752	3.507
	This study	3.670	-1.322	-0.988	0.557
R^2	Eq. 15	0.005	0.986	0.986	0.148
	This study	0.984	0.958	0.959	0.980

Table 9
Statistical parameters of this study compared with other models for ethylene/O_2 mixtures.

		Ref. [2]	Ref. [21]	Total data
AARD%	Eq. 16	64.647	40.935	41.486
	Eq. 17	99.999	99.999	99.999
	Eq. 19	14.046	47.999	41.702

	Eq. 20	35.243	68.831	62.454
	This study	29.261	29.539	29.485
ARD%	Eq. 16	-64.166	-23.972	-25.883
	Eq. 17	99.999	99.999	99.999
	Eq. 19	0.846	34.046	29.073
	Eq. 20	-35.065	10.169	-7.413
	This study	-11.797	18.466	12.594
R^2	Eq. 16	0.938	0.832	0.837
	Eq. 17	0.899	-0.453	-0.412
	Eq. 19	0.996	0.253	0.285
	Eq. 20	0.987	0.364	0.389
	This study	0.989	0.946	0.947

These tables show that the proposed correlation is more accurate than other models for total data in each fuel. The differences between AARD% of our model and previous correlations are considerable. For each fuel, the AARD% value of proposed model is the lowest. Only in case of kerosene fuel, the AARD% of new model is a little higher, which is negligible. Comparing the R^2 value of total data for each fuel, it was shown that the proposed model had the value closer to 1. This means that, the calculated ignition delay time data are in agreement with the experimental ones. According to Tables 3-9, it is acknowledged that the available models in literature can predict only their data well. However, for the same fuel data from other literature, they cannot predict accurately. The new model is capable of correlating the total data for each fuel in different references well.

To compare the accuracy of the presented empirical model, calculated ignition delay time data for every substance versus corresponded values in data bank have been drawn in Figs. 1-4. The aggregation of data around the bisector shows that the calculated ignition delay time data are close to experimental values in different ranges. For heavy hydrocarbon fuels (decane, kerosene, Jet-A), some data do not follow this condition. It can be related to measurement errors in experimental tests. In addition, it can be resulted because these fuels are a complex mixture of several hundreds of hydrocarbons including alkanes, cycloalkanes, aromatics and polycyclic compounds, and the detailed composition of them generally varies with each source.

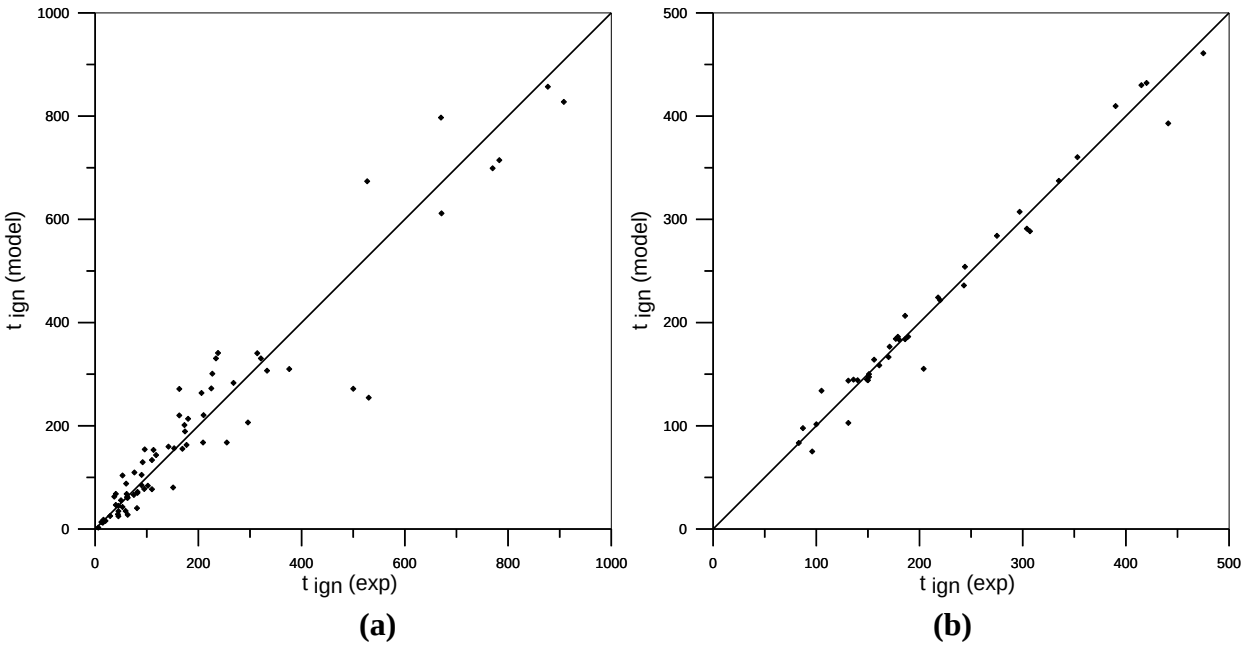

Figure 1. Accuracy of presented model versus sources data: (a) methane (b) butane.

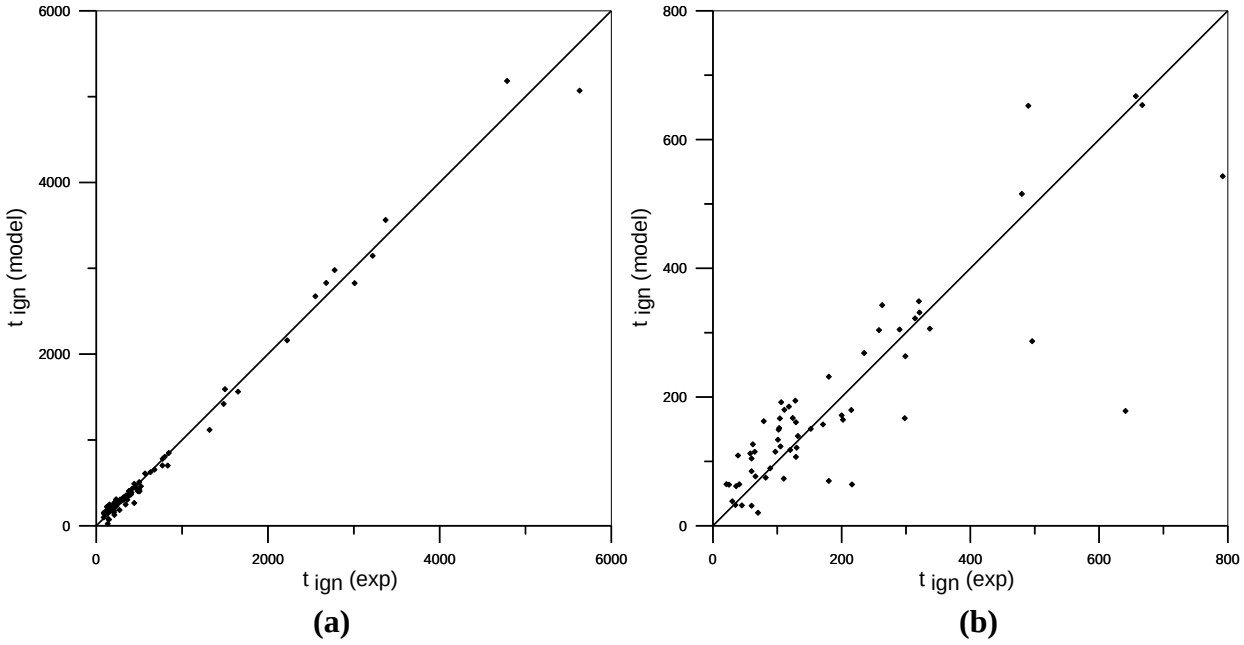

Figure 2. Accuracy of presented model versus sources data: (a) heptanes (b) decane.

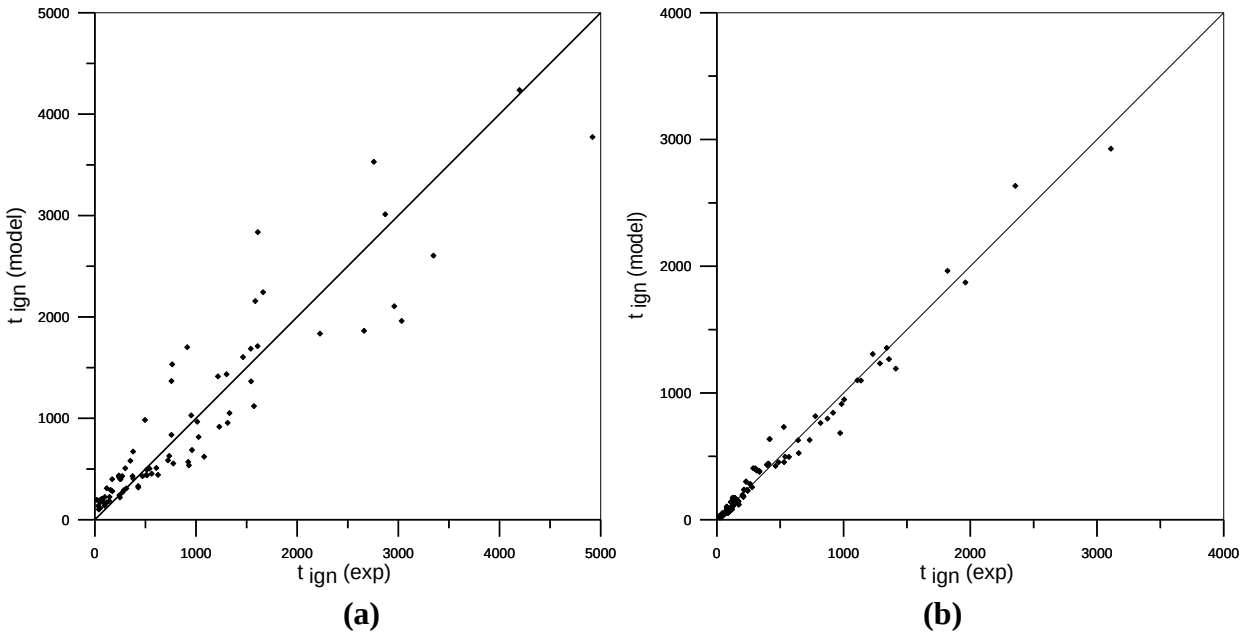

Figure 3. Accuracy of presented model versus sources data: (a) kerosene (b) Jet-A.

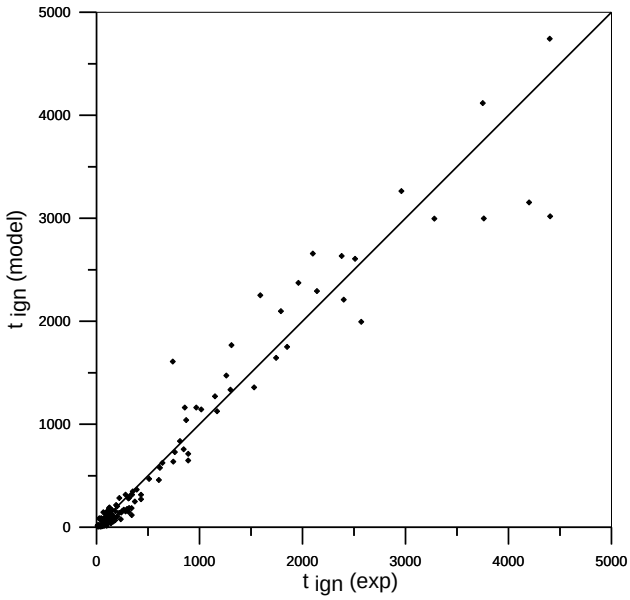

Figure 4. Accuracy of presented model versus sources data: ethylene.

4. Conclusions

In the case of methane, butane, heptane, decane, kerosene, Jet-A and ethylene fuels, a new predictive correlation for the ignition delay time as a function of pressure, number of carbon atoms, mixture equivalence ratio, fuel mole fraction and temperature was recommended. This model was derived from data sources reported in literature. It was found that undesirable prediction deviations were obtained using previous models for all data in each fuel. The new correlation with constant parameters for each substance generally gave good prediction accuracy

relative to other models. To validate the proposed model, the ignition delay time data for each fuel have been examined and an overall average absolute relative deviation was calculated for each substance. The amount of AARD% in our model was much less than other correlations for the fuels and R^2 value of total data is closer to 1. It was recognized that the available models in literature could predict only their data well. However, for the same fuel data from other literature, they could not predict accurately. The new model is able to correlate the total data for each fuel in different references well.

Nomenclature

AARD%	Average absolute relative deviation percentage
ARD%	Average relative deviation
E	Activation energy (cal/mol)
φ	Equivalence ratio
τ	Ignition delay time
μs	Microsecond

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