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T.T. Al-Shemmeri, S. Oberweis

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PII: S1359-4311(11)00090-1
DOI: 10.1016/j.applthermaleng.2011.02.010
Reference: ATE 3420

To appear in: Applied Thermal Engineering

Received Date: 8 November 2010
Revised Date: 1 February 2011
Accepted Date: 9 February 2011

Please cite this article as: T.T. Al-Shemmeri, S. Oberweis. Correlation of the NO\textsubscript{x} emission and exhaust gas temperature for biodiesel, Applied Thermal Engineering (2011), doi: 10.1016/j.applthermaleng.2011.02.010

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Correlation of the NO$_x$ emission and exhaust gas temperature for biodiesel

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Abstract

This paper presents an algorithm which correlate Nitrogen oxides emitted and the combustion flame temperature during the combustion of biodiesel. An iterative process is used to determine the flame temperature taking into accounts the phenomenon of dissociation. The results of the algorithm are presented for different and air-to-fuel ratios. These predicted results are compared with laboratory tests conducted in the present study on a stationary diesel engine run on different blends of biodiesel. Within the range of tests carried out, the NO$_x$ emissions from biodiesel and its blends proved to be higher than those of petro-diesel fuel. Furthermore, in this study a strong correlation was found relating the NO$_x$ emissions and the flame temperature.

Keywords: biodiesel blends, dissociation, NO$_x$ emission.
### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
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</thead>
<tbody>
<tr>
<td>$B$</td>
<td>Blend ratio</td>
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<tr>
<td>$G$</td>
<td>Gibbs function</td>
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<td>$GHG$</td>
<td>Greenhouse gas emission</td>
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<tr>
<td>$H_o$</td>
<td>Enthalpy of formation at STP (kJ/kmol)</td>
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<tr>
<td>$HHV$</td>
<td>Higher Heating Value (MJ/kg)</td>
</tr>
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<td>Kinetic energy (J)</td>
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<tr>
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<td>Dissociation equilibrium constant</td>
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<tr>
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<td>Number of moles</td>
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<td>Potential energy (J)</td>
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<td>Pressure (Pa)</td>
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<td>Heat transfer (W)</td>
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<td>Entropy (kJ/kgK)</td>
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<tr>
<td>$TE$</td>
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<tr>
<td>$VOC$</td>
<td>Volatile Organic Compounds</td>
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<td>$W$</td>
<td>Work transfer (J)</td>
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#### Greek letters

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<th>Definition</th>
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<tr>
<td>$\Delta$</td>
<td>Difference, change</td>
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<td>$\nu$</td>
<td>Mole fraction</td>
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#### Subscripts

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<td>$P$</td>
<td>Products of reaction</td>
</tr>
<tr>
<td>$R$</td>
<td>Reactants</td>
</tr>
<tr>
<td>$o$</td>
<td>Standard conditions</td>
</tr>
<tr>
<td>$s$</td>
<td>Stoichiometric</td>
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</table>
1. Introduction

The worryingly rapid depletion of fossil fuel is demanding an urgent need to carry out research work regarding alternative fuels. It is uncertain how much oil and gas resources are available or remain to be discovered. Starting in 2008 the Renewable Transport Fuel Obligation in the UK will "place an obligation on fuel suppliers to ensure that a certain percentage of their aggregate sales is made up of biofuels. [1]. Fossil fuels accounted for 85% of world’s primary energy supply and over 94% of energy for transportation. The production of use of fossil fuels contribute to 80% of anthropogenic GHG emissions worldwide and fossil fuel power generation currently accounts for over one third of global annual carbon dioxide emissions [2].

In 1999, European countries signed a protocol to abate acidification, eutrophication and ground-level ozone in Gothenburg [3]. The protocol sets emission ceilings for 2010 for four pollutants: sulphur, NO\textsubscript{x}, VOCs and ammonia. These ceilings were negotiated on the basis of scientific assessments of pollution effects and abatement options. Parties whose emissions have a more severe environmental or health impact and whose emissions are relatively cheap to reduce will have to make the biggest cuts. Once the protocol is fully implemented, Europe’s sulphur emissions should be cut by at least 63%, its NO\textsubscript{x} emissions by 41%, its VOC emissions by 40% and its ammonia emissions by 17% compared to 1990. The protocol also sets tight limit values for specific emission sources (e.g. combustion plant, electricity production, dry cleaning, cars and lorries) and requires best available techniques to be used to keep emissions down. Only recently, has the European council decided to commit itself to increasing the percentage of renewable energies of the total primary energy consumption to 20% by 2020 inside the EU [4]. In recent years, the reduction in sulphur content is the most notable restriction. Due to
diminishing fossil fuel reserves and more and more stringent emission limits, biodiesel has yet again become popular [5].

The term biodiesel commonly refers to fatty acid methyl esters made from vegetable oils or animal fats, whose properties are good enough to be used in diesel engines. Since vegetables have cetane numbers close to that of diesel fuel, they can be used in existing compression ignition engines with little or no modifications [6-8] and the production of biodiesel is currently regulated by standard EN-14214 [9-10] in Europe. Several studies have investigated the effects on emissions due to blending diesel with biodiesel [11, 12]. The formation of mono nitrogen oxides can lead either to an increase or decrease due to blending [13, 14]. These papers show that in general pre 1997 diesel engines have an increase in NO\textsubscript{x} emission with increased biodiesel percentage due to problems with injection timings, which is after all designed in accordance with the fuels viscosity [15]. Research papers presenting results of diesel engine emissions from biodiesel often ignore some of the basic properties of the biodiesel used, which makes it difficult to determine whether its quality has some effect or not. Lapuerta et al. [16] presented a review showing that most published articles reported that biodiesel present an increase in NO\textsubscript{x} compared with petro-diesel and only about 5% of the articles show a decrease. The present study presents the reader with an algorithm to estimate Nitrogen oxides formation and the combustion flame temperature for biodiesel depending on its chemical composition. The theoretical results will be compared to laboratory testing of a stationary diesel engine run on different blends of biodiesel.
2. Thermodynamic Analysis of the Combustion of Biodiesel in CI Engines

2.1 Stoechiometric relations

Much of the analysis in this section was taken from reference [17]. The first law of Thermodynamics is based on the principle of energy conservation within a system. This concept, when applied to a typical engine cylinder, can be considered as an open system, for which the Steady Flow Energy Equation (SFEE):

\[ Q - W = \Delta H + \Delta KE + \Delta PE \]  

For a typical combustion inside the cylinder, the flow process is considered adiabatic (Q=0), with no work transfer and the changes of kinetic and potential energy can be neglected; the above equation becomes:

\[ \Sigma \Delta H_{p} = \Sigma \Delta H_{r} \]  

The enthalpies of products and reactants are functions of the temperature which are tabulated in standard thermodynamics textbooks. In order to evaluate the flame temperature, an iterative process is employed with the key to find that temperature (from the tables, enthalpies of combustion versus temperature) until equation (2) is satisfied.

In order to determine the different properties of the combustion in question, the chemical formula of the fuel and the Stoechiometric equation are required. Consider the following general hydrocarbon fuel combustion:

\[
\begin{align*}
[Fuel] & + \hspace{1cm} [Air] = \text{Products of combustion} \\
[R_1C] + [R_2H] + [R_3O] + [R_4N] + [R_5S] + [R_6O_2] & \rightarrow [P_1CO_2] + [P_2H_2O] + [P_3NO_2] + [P_4SO_2] + [P_5N_2] + [P_6O_2]
\end{align*}
\]
The Stoechiometric air-fuel ratio (A/F) in which \( P_6 \) is zero, can be derived from the equation of complete combustion equation (3):

\[
\left( \frac{A}{F} \right)_i = \frac{R_6[32 + (3.76 \times 28)]}{12R_1 + R_2 + 16R_3 + 14R_4 + 32R_5}
\] (4)

\[
(\%){NO_2} = \frac{P_5}{P_1 + P_2 + P_3 + P_4 + P_5 + P_6}
\] (5)

In the presence of excess air equation (3) can be rewritten

\[
C_{R1}H_{R2}O_{R3}N_{R4}S_{R5} + (1 + x)R_6[O_2 + 3.76N_2] 
\rightarrow P_1CO_2 + P_2H_2O + P_3NO_2 + P_4SO_2 + \left[\left(P_5 + x\right)N_2\right] + \left[\left(P_6 + x\right)O_2\right]
\] (3a)

2.2 Dissociation

During the combustion process of any fuel, several chemical reactions take place at the same time. Take for example the combustion of pure carbon in oxygen:

\[
C + O_2 \leftrightarrow CO_2 + TE
\] (6)

The energy released (TE) during the combustion may under certain conditions be enough for the carbon dioxide to undergo the reverse reaction. This reaction is endothermic (absorbing energy from the products) and hence will reduce the temperature of the products. Dissociation of the individual species of the bio-diesel implies that the combustion process of some of the reactions is reversible; hence some of the energy indicated in equation (6) will be lost. The reaction equation (7) will adjust itself to an equilibrium state. This phenomenon is known as dissociation and it is
responsible for lowering the combustion temperature. At some temperature $T$, a fraction of $\nu$ moles of any species among the products dissociate. For the C-H-O-N system, the complete chemical equilibrium scheme proposed by Way [20] is used. The following reverse reactions are included in the calculations:

\[
\begin{align*}
CO + \frac{1}{2}O_2 & \rightleftharpoons (1 - \nu)CO_2 \\
H + H & \rightleftharpoons (1 - \nu)H_2 \\
O + O & \rightleftharpoons (1 - \nu)O_2 \\
N + N & \rightleftharpoons (1 - \nu)N_2 \\
\frac{1}{2}O_2 + \frac{1}{2}N_2 & \rightleftharpoons (1 - \nu)NO \\
\frac{1}{2}H_2 + OH & \rightleftharpoons (1 - \nu)H_2O \\
H_2 + \frac{1}{2}O_2 & \rightleftharpoons (1 - \nu)H_2O
\end{align*}
\]

Generally, the dissociated reaction takes the following form:

\[
\Delta \mu = \sum \nu_i \mu_i R \rightleftharpoons \sum \nu_p \mu_p R_p
\]

A general equilibrium constant for any reverse reaction is given by:

\[
K_p = \frac{N_p \nu_p N_r \nu_r}{N_0 \nu_0 N_r \nu_r} \left( \frac{1}{N_{tot}} \right)^\Delta
\]

In order to determine $K_p$, the Gibbs function is commonly used:

\[
\Delta G = V\Delta P - S\Delta T
\]

At equilibrium, the temperature is constant, ($\Delta T = 0$) and substitute $V = nRT / P$ for ideal gas, then the above equation becomes:
\[ \Delta G = N \, R \, T \, \Delta \frac{P}{\bar{P}} \]  \hspace{1cm} (17)

Integrating between state 1 and 2:

\[ G_2 - G_1 = \bar{R} \, T \, \ln \frac{\bar{P}^2}{P^1} \]  \hspace{1cm} (18)

For a reversible reaction with two reactants and two products the above equation becomes:

\[ G_p - G_R = R \, T \, \ln \left[ \frac{P_{r_1} \, P_{r_2}}{P_{p_1} \, P_{p_2}} \right] \]  \hspace{1cm} (19)

The square bracket on the RHS of equation 19 is known as the equilibrium constant \( K_p \); values for \( \ln[K_p] \) are tabulated in Table 1.

Now equation (15) can be solved for \( \upsilon \) for each individual constituent, hence determines the dissociated form of combustion.

The Stoechiometric reaction for a typical fuel is given by equation (3). As this does not take into account dissociation of the products, it needs to be changed to reflect the molecules occurring in the products.

3. Algorithm Structure

The combustion theory outlined in section 2 is used to develop a computer model to investigate the emission from the combustion of Biodiesel, Fig. 1.

The model processes the calculations in the following stages, which can be grouped in two main parts:

A. Stoechiometric calculations – this includes:

   A1. Input fuel composition, and fuel properties

   A1. Determine the chemically balanced equation of combustion (equation 3)
A3. Starting with a guess value of temperature as 298K, iterate (increasing this
temperature by one degree Celsius each step) till equation 2 is satisfied, to determine the
adiabatic flame temperature.

B. Dissociation calculations – this includes:
B1. Look up Kp values in table 1, based on the adiabatic flame temperature
B3. Determine the actual products of combustion, based on the results in B2.
B4. Starting with a guess value of temperature as 298K, iterate (increasing this
temperature by one degree Celsius each step) till equation 2 is satisfied, to determine the
actual flame temperature.
B5. Loop B1-B4, with starting point the actual flame temperature until it converges, i.e.
the new actual flame temperature minus old actual flame temperature equals less or
equal than 0.1 K.
B6. Depending on A/F ratios, step A2 can be modified to account for excess air. A re-
run of steps A1 – B5 is repeated.

4. Experimental engine and test facility

4.1 The test rig
The test rig, shown in Fig. 2, consists of the prime mover which is a Lister-Petter diesel
engine coupled to an electrical generator (engine specifications are found in table 2) and
a heat exchanger. In addition, instruments include: a data logger a personal computer for
logging and processing the data; an air box and a U-tube manometer to measure the air
intake of the engine; a fuel flow rate measurement system to measure the fuel
consumption; electrical voltage and current meters to measure the power output of the
engine; K-type thermocouples to measure air temperature, exhaust gas temperature and the temperatures at different points on the heat exchanger; and finally emission analysers to measure the composition of the exhaust gas, the specifications for the emission analysers are found in table 3.

4.2 Fuel samples and properties

The conventional petro-diesel fuel was supplied by ESSO UK part of the EXXON Mobil Corporation and represents the typical, British automotive, low sulphur (0.005%) diesel fuel. The biodiesel used in this study conforms to the European standards regulating the manufacture of biodiesel, EN14214 [9]. The ultimate analysis for the two fuels used in the experiment is obtained from the manufacturers presented in Table 4. The fuels were blended in the laboratory and to prevent mixing of different blends in the tank, the engine was left running dry at the end of each test group. Bleeding the engine was necessary prior to every new test run with a new fuel blend. The blends used in this experiment are B25, B50 and B75; the letter B assigned for Biodiesel followed by the percentage by volume blended. Additional runs were carried out with pure petro-diesel and pure biodiesel. Therefore there are 5 fuel samples tested in this study B0, B25, B50, B75 and B100.

4.3 Parameters tested and experimental procedure

A series of tests were conducted using each of the above 5 fuel blends, with the engine working at a speed of 1500 rpm and five engine loads ranging from no-load to full-load. In each test, volumetric fuel consumption rate, exhaust gas temperature, exhaust regulated gas emissions such as nitrogen oxides, carbon monoxide, carbon dioxide and total unburned hydrocarbons are measured. The experimental work started with preliminary investigation of the engine running on neat diesel fuel, in order to determine
the engine’s operating characteristics and exhaust emission levels, constituting a ‘baseline’ that is compared with the corresponding cases when using the subsequent fuel blends. The same procedure was repeated for each fuel blend by keeping the same operating conditions. For every fuel change, the engine was run ‘dry’ and after the ‘bleeding’ process, was left to run for thirty minutes to stabilise at its new condition. Each test was repeated further 3 times to reduce experimental error.

5. Results and Discussion

The objective of this study is to establish a computer program to estimate exhaust gas emission from hydrocarbons and its flame temperature. The accuracy of this program is compared to the experimental test results of biodiesel blends in a stationary diesel engine.

5.1 Exhaust gas temperature

The variation of exhaust gas temperature with respect to the load for B50 blend is shown in Fig. 3. The error between measured and predicted temperatures for B50 was in the range of 4-9.5% and showed an increasing trend with increased engine loads. This error is most likely due to the fact that software does not take into account left over heat from previous operations. As the engine was not cooled down for operating reasons prior to increasing the engine load. It can be seen on Fig. 3 that the measured temperature is indeed higher than the predicted one. The difference between the predicted temperatures and published data about experiments carried out under similar conditions lies within the range of 0-4.5%.

5.2 Nitrogen oxides emission (NOx)

The variation of NOx emission with respect to different blending at no-load is shown in Figures 4 and 5. The difference between predicted and measured emissions for Nitrogen
oxides is in the range of 0.7-5%. The predicted emission of NO\textsubscript{x} is based on the
dissociation of Oxygen and Nitrogen in the exhaust gases and the resulting temperature.
As the NO\textsubscript{x} increases directly with the degree of blending and as the latter is directly
proportional to the temperature, this proves that the most important factor for the
emission of NO\textsubscript{x} is the combustion temperature. Fig. 4 shows that within the range of
the tests, the NO\textsubscript{x} emissions from biodiesel and its blends are higher than those of diesel
fuel. The difference of the published (a summary of 38 studies) results has a maximum
of 6% for standard diesel. The majority of studies in this publication are based on
American fuels and, hence might have some slight differences with the molecular
structure compared to European type petro diesel. Fig. 6. shows indeed a linear
correlation between exhaust gas temperature and NO\textsubscript{x} emissions for predicted values for
B100 blend. The relationship between the NO\textsubscript{x} emitted and the exhaust temperature was
correlated, and the fitted equation was found to be:

\[ \text{NO}_x = 0.3061 \times T - 206.85 \]

The fitted data were excellent, the R-squared of the fit was found to be: \( R^2 = 0.999 \).

6. Conclusion

In this study, an algorithm to estimate Nitrogen oxides formation and the resulting
flame temperature for biodiesel depending on its chemical composition was introduced.
A computational program was developed for simulating the combustion of biodiesel and
its blends depending on their chemical composition and the resulting flame temperature.
The findings were compared to laboratory testing and published results from similar
experiments. The main conclusions from the current study can be summarised as
follows.

6.1 NO\textsubscript{x} for each test (Fig. 4 & Fig. 5) varied linearly with blend. The same
behaviour was observed for all 5 loads.
6.2 The flame temperature variation (Fig. 3) followed a linear relation and there was a close relation between measured, predicted and published data. The same behaviour was found for each blend over the range of 5 loads tested.

6.3 In an attempt to correlate the variation of flame temperature and its corresponding NO\textsubscript{x} emission (Fig. 6) it was found that a linear relationship existed, although it has been suspected, these results was a pleasant finding.

6.4 The present algorithm was validated further against some published data [21] and it was found in good agreement with their results.

6.5 The present algorithm, although developed initially to correlate the biodiesel blends combustion, currently the authors are working on extending it for biomass combustion.

References


Research Highlights for publication Ref. ATE_3420

- A correlation of Nitrogen oxides & the combustion flame temperature of biodiesel.
- An iterative process is used to determine the flame temperature with dissociation.
- The present algorithm can be applied to biomass combustion.
Captions for Tables

Table 1  The natural logarithm of $K_p$ for various products [17]
Table 2  Test engine specifications.
Table 3  Specifications of exhaust gas analysers used in measurements in present study.
Table 4  Ultimate analysis and HHV of the fuels used in the present study
Table 1 - The natural logarithm of $K_p$ for various products [17]

<table>
<thead>
<tr>
<th>Temperature [K]</th>
<th>$H_2 = 2H$</th>
<th>$O_2 = 2O$</th>
<th>$N_2 = 2N$</th>
<th>$H_2O = 1/2H_2 + 1/2O_2$</th>
<th>$H_2O = 1/2H_2 + OH$</th>
<th>$CO_2 = CO + 1/2O_2$</th>
<th>$1/2N_2 + 1/2O_2 = NO$</th>
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<tr>
<td>298</td>
<td>-164.01</td>
<td>-186.98</td>
<td>-367.48</td>
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<td>-106.21</td>
<td>-103.76</td>
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<td>-119.94</td>
<td>-240.63</td>
<td>-59.60</td>
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<td>1200</td>
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<td>-80.01</td>
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<td>-2.11</td>
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Table 2 - Test engine specifications

<table>
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<tr>
<th>Parameters</th>
<th>Value</th>
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<tr>
<td>Generator voltage [V]</td>
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<tr>
<td>Full load current [A]</td>
<td>10</td>
</tr>
<tr>
<td>Gen-Set power [kVA]</td>
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<tr>
<td>No of cylinders</td>
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<tr>
<td>Swept volume [cc]</td>
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<tr>
<td>Compression ratio</td>
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<tr>
<td>Bore [mm]</td>
<td>95.3</td>
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<tr>
<td>Stroke [mm]</td>
<td>88.9</td>
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<td>Speed fixed [rpm]</td>
<td>1500</td>
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Manufacturer - Lister-Petter; Model - TS2
Table 3 - Specifications of exhaust gas analysers used in measurements in present study

<table>
<thead>
<tr>
<th>Object of measurement</th>
<th>CO</th>
<th>CO₂</th>
<th>NOₓ</th>
<th>O₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Range of measurement</td>
<td>0 ~ 3 %</td>
<td>0 ~ 20 %</td>
<td>0 ~ 10,000 ppm</td>
<td>0 ~ 100 %</td>
</tr>
<tr>
<td>Resolution</td>
<td>0.002 %</td>
<td>0.1 %</td>
<td>50 ppm</td>
<td>0.1 %</td>
</tr>
<tr>
<td>Accuracy</td>
<td>1 ~ 2 %</td>
<td>1 ~ 2 %</td>
<td>1 ~ 2 %</td>
<td>0.05 %</td>
</tr>
</tbody>
</table>

Manufacturer: (CO, CO₂, NOₓ) : Analytical development Co Ltd (ADC) ;

(O₂): Servomex Ltd.
Table 4 - Ultimate analysis and HHV of the fuels used in the present study

<table>
<thead>
<tr>
<th></th>
<th>B0</th>
<th>B25 (^2)</th>
<th>B50 (^2)</th>
<th>B75 (^2)</th>
<th>B100</th>
</tr>
</thead>
<tbody>
<tr>
<td>C wt%</td>
<td>86.400</td>
<td>84.075</td>
<td>81.750</td>
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<td>77.100</td>
</tr>
<tr>
<td>H</td>
<td>12.700</td>
<td>12.425</td>
<td>12.150</td>
<td>11.875</td>
<td>11.600</td>
</tr>
<tr>
<td>O</td>
<td>0.040</td>
<td>2.730</td>
<td>5.420</td>
<td>8.110</td>
<td>10.800</td>
</tr>
<tr>
<td>N</td>
<td>0.085</td>
<td>0.189</td>
<td>0.293</td>
<td>0.396</td>
<td>0.500</td>
</tr>
<tr>
<td>S</td>
<td>0.005</td>
<td>0.004</td>
<td>0.003</td>
<td>0.001</td>
<td>0.000</td>
</tr>
<tr>
<td>HHV (^1) [MJ/kg]</td>
<td>42.30</td>
<td>40.98</td>
<td>39.65</td>
<td>38.33</td>
<td>37.00</td>
</tr>
</tbody>
</table>

1: www.iea.org (accessed 18.02.2010),
2: computed
3. columns B0 & B100 taken from reference [22]
Captions for Figures

Fig. 1  Computer program flowchart highlighting iteration process
Fig. 2  Schematic diagram of the experimental setup
Fig. 3  Measured, predicted and published temperatures at half load
Fig. 4  Measured NO\textsubscript{x} results for different engine loads and fuel blends
Fig. 5  Predicted NO\textsubscript{x} results for different engine loads and fuel blends
Fig. 6  Correlation for predicted NO\textsubscript{x} emission and exhaust gas temperature for B100
Fig. 1 Computer program flowchart highlighting iteration process
Fig. 2 Schematic diagram of the experimental setup
Fig. 3 Measured, predicted and published temperatures at half load.
Fig. 4 Measured NOx results for different engine loads and fuel blends
Fig. 5 Predicted NOx results for different engine loads and fuel blends

\[ \text{NO}_x = 0.1084 \times \% \text{Blend} + 5.771 \]

\[ R^2 = 0.999 \quad (\text{full load}) \]
Fig. 6 Correlation of predicted NOx emission and exhaust gas temperatures for B100.