

Thermodynamic Properties of Gas Generated by Rapeseed Methyl Ester-Air Combustion Under Fuel-Lean Conditions

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ABSTRACT

Biodiesel in the form of rapeseed methyl ester (RME) has been widely used for transportation as a partial (or complete) substitute fuel for compression ignition (diesel) engines. The properties of gas generated by the combustion of RME and air under fuel-lean conditions were studied. A simple analytical model was used to determine the burned gas composition. Firstly, the effects of relative air/fuel ratios on flue gas composition were studied theoretically, without exhaust gas recirculation (EGR). As the relative air/fuel ratios increased, carbon dioxide and water vapor decreased, while oxygen increased in the burned gases. The mass and molecular weight of the burned gas reduced with increasing relative air/fuel ratios. Secondly, the thermodynamic properties (i.e. specific heat, specific heat ratio, enthalpy and entropy) of the burned gas mixture, as they were affected by the relative air/fuel ratio, were calculated based upon the element concentrations of individual component in the chemical equilibrium state. The thermodynamic properties of RME-air combustion gas were plotted against relative air/fuel ratios under fuel-lean conditions and were compared to those from conventional diesel fuels. Under certain conditions, good agreement was found. This research produced primary data for further calculations regarding the thermochemistry of biodiesel-air combustion analysis.

Key words: thermodynamic properties, biodiesel, rapeseed methyl ester, combustion, lean fuel

INTRODUCTION

The use of biodiesel as a part-substitute for diesel fuel has been promoted as a means of reducing petroleum fuel consumption. As one among other alternative bio-fuels, biodiesel's properties are comparable to those of conventional diesel fuel, except for the favorable near-zero sulfur and aromatic contents (see Table 1 for a fuel property comparison). In addition, promotions to use other renewable bio-fuels in transport (Directive 2003/30/EC, 2003) and environmental

concerns on harmful emissions are probably the main reasons for instigating the use of biodiesel as an alternative fuel. In Europe and other regions, vehicles with un-modified diesel engines are mostly capable of using up to 5% by volume of biodiesel in the blend directly, without any problem (Directive 2003/30/EC, 2003). Additionally, there is an ambitious European Union program described in a European Commission Green Paper (CEC, 2000), which has set a target of 20% substitution of conventional fuels by alternative fuels in the road transport

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sector, by the year 2020.

Biodiesel in the form of methyl or ethyl ester is composed of oxygenated compounds, defined as mono alkyl esters of long-chain fatty acids derived from lipid feedstock, such as vegetable oils and animal fats (Graboski and McCormick, 1998). Pure oils are not suitable for diesel engines, as they were reported to cause a variety of problems, including long-term engine deposits, injector plugging or lube oil gelling (Kalam and Masjuki, 2005). Biodiesel is produced through the transesterification of pure oils, while methyl ester-based biodiesel production has been shown to have a higher yield than ethyl ester production (Sanli and Canakci, 2008). In the transesterification process, renewable feedstock in the form of triglycerides reacts with methanol in the presence of a catalyst to yield methyl ester and by-products (Kinast, 2003). Due to its cost effectiveness and reaction stability, a base catalyst (for example, sodium hydroxide) is usually used (Graboski and McCormick, 1998). The by-products comprise: glycerol, water, traces of methanol, un-reacted triglycerides and catalyst (Babu and Devaradjane, 2003). Generally, the

typical heating value for biodiesel is lower than that of conventional diesel fuel (McCormick, 2003); example values can be seen in Table 1. As a result, a greater amount of fuel is required in order to maintain the same engine brake torque. From a stock engine, without timing changes or an exhaust catalyst, biodiesel and its blends reduce most engine emissions that consist of total unburned hydrocarbons (HC), carbon monoxide (CO), and particulate matter (PM), compared to conventional diesel engines, while the only regulated emission shown to increase consistently with biodiesel is nitrogen oxides (NO_x) (Graboski *et al.*, 2003). However, an exhaust gas recirculation (EGR) technique can mitigate the increasing NO_x (Ladommatos *et al.*, 1998). EGR, as an additional diluent to fresh air, reduced NO_x emissions by lowering the radical species concentration presented during the combustion process (Montgomery and Reitz, 2001). Some other unregulated emissions, such as polycyclic aromatic hydrocarbons (PAH), nitro PAH compounds and some toxic reactive HC species, are significantly reduced by the use of biodiesel (Sharp *et al.*, 2000).

Rapeseed is one of the main oil-bearing

Table 1 Fuel properties.

Fuel analysis	Rapeseed methyl ester (RME)	Ultra low sulfur diesel (ULSD)
Cetane number	54.7	53.9
Density at 15°C (kg/m ³)	883.7	827.1
Viscosity at 40°C (cSt)	4.478	2.467
Lower calorific value (MJ/kg)	39.0	42.7
Sulfur (mg/kg)	5	46
Mono-aromatics (% wt)	21.0	<0.1
Di-aromatics (% wt)	3.1	<0.1
Molecular weight	296	209
C (% wt)	77.2	86.5
H (% wt)	12.0	13.5
O (% wt)	10.8	-
Empirical formula	C _{19.03} H _{35.17} O ₂	C _{15.05} H _{27.94}

Source: Chuepeng *et al.* (2007)

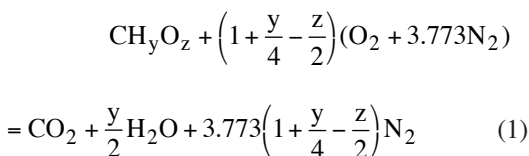
plants widely used as a feedstock for producing biodiesel in Europe and some other parts of the world (Demirbas, 2007). Previous published work has detailed studies concerning the use of rapeseed methyl ester (RME) in compression ignition (CI) engines; some extensive aspects based up on experimental results are reviewed here. Tsolakis *et al.* (2007) studied the engine performance and emissions of a diesel engine operating on diesel-RME blends with EGR. Chuepeng *et al.* (2007) determined combustion characteristics (including the heat release pattern and ignition delay) in order to assess quantitative impacts of emissions from biodiesel blends containing a high proportion of RME. Chuepeng *et al.* (2008a) investigated the number and mass-size distribution of nanoparticles in the exhaust gas of diesel engines fuelled with RME blends. Considering exhaust gas after-treatment, Chuepeng *et al.* (2008b) produced on-board hydrogen-rich gas as a reformed EGR by an exhaust-gas-assisted fuel-reforming technique, using biodiesel-blended fuels. This method has been shown to reduce NO_x and smoke emissions simultaneously.

Thermodynamic data of the burned gas from isooctane (C_8H_{18}) and other C_nH_{2n} fuels combusted with air have been gathered in Heywood (1988). However, for other oxygenated fuels, studies on biodiesel combustion by both experimentation and modeling require crucial thermodynamic data for calculating and explaining some of the phenomena that occur. In particular, rapeseed methyl ester-air combustion data are not readily available. Therefore, the main aim of this paper was to present calculations on the thermodynamic properties of a burned gas mixture under fuel-lean conditions, representing a real diesel engine load under operating conditions. Prior to the calculations, a simple analytical model was used to determine the burned gas composition. The data was then compared with those of conventional diesel fuel under certain conditions.

MATERIALS AND METHODS

Burned gas composition determination

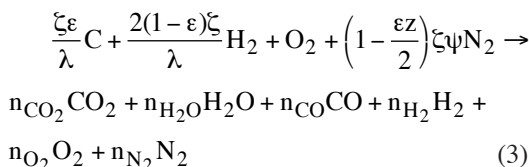
The proposed analytical model for calculating the burned gas fraction was originally given in Heywood (1988) for hydrocarbon fuel combustion and has been applied here to an oxygenated fuel. For rapeseed methyl ester (empirical formula CH_yO_z), the composition of the burned gas fraction can be theoretically calculated using the stoichiometric combustion Equation 1:



In such circumstances, combustion can occur under fuel-rich or fuel-lean conditions. Therefore, the relative air/fuel ratio (λ) defined in Equation 2 is used:

$$\lambda = \frac{(\text{A}/\text{F})_{\text{actual}}}{(\text{A}/\text{F})_{\text{stoichiometric}}} \quad (2)$$

where, $(\text{A}/\text{F})_{\text{actual}}$ is the air-to-fuel ratio on a mass basis used in real combustion and $(\text{A}/\text{F})_{\text{stoichiometric}}$ is the chemically correct or theoretical proportion of air and fuel on a mass basis. Combustion Equation 1 can be written per mole of oxygen (O_2) using Equation 3:



$$\text{where, } \xi = \frac{2}{2 - \varepsilon z \left(1 - \frac{1}{\lambda}\right)}$$

ψ ; -e molar N/O ratio (3.773 for air)

$\varepsilon = 4/(4+y)$

n_i = moles of species i per mole O_2 reactant

The n_i are determined using the assumption for fuel-lean and stoichiometric mixtures ($\lambda \geq 1$) in a typical diesel engine operation, that amounts of carbon monoxide (CO) and hydrogen (H_2) are minor and can be neglected. Based on the elemental balance and the above assumption, the n_i can be obtained (Table 2). However, this simple analytical method does have some limitations; the regulated products of combustion (such as NO_x , total unburned HC and CO) and other unregulated harmful emissions (PAH and nitro PAH compounds, among others) cannot be determined.

The mole fractions, \tilde{x} , are calculated by Equation 4:

$$\tilde{x}_i = \frac{n_i}{n_b} \quad (4)$$

where, $n_b = \sum_i n_i$ is given in Table 2.

The mass of the burned gas per mole O_2 in the mixture, m_b , is (Equation 5):

$$m_b = 32 + \frac{4(2\varepsilon + 1)}{\lambda} + 28.16\psi \quad (5)$$

The molecular weight of the burned gas at low temperature (<1700 K), M_b , can be therefore

calculated by Equation 6:

$$M_b = \frac{m_b}{n_b} \quad (6)$$

Calculation of thermodynamic properties of the burned gas

The approach concisely given in Heywood (1998) and applied here is based on the NASA equilibrium program with JANAF (Joint Army Navy Air Force) table thermodynamic data (Gordon and McBride, 1971). A polynomial curve can be fitted to the thermodynamic data for each species in the mixture, on the assumption that the burned gas is in an equilibrium gaseous phase.

The specific heat at constant pressure for each species i ($\tilde{c}_{p,i}$) in its standard state at temperature T (K) is approximated by Equation 7:

$$\frac{\tilde{c}_{p,i}}{R} = a_{i1} + a_{i2}T + a_{i3}T^2 + a_{i4}T^3 + a_{i5}T^4 \quad (7)$$

The standard state enthalpy of species i (\tilde{h}_i) is given by Equation 8:

$$\frac{\tilde{h}_i}{RT} = a_{i1} + \frac{a_{i2}}{2}T + \frac{a_{i3}}{3}T^2 + \frac{a_{i4}}{4}T^3 + \frac{a_{i5}}{5}T^4 + \frac{a_{i6}}{T} \quad (8)$$

The standard state entropy of species i (\tilde{s}_i) at temperature T (K) is provided by Equation 9:

$$\frac{\tilde{s}_i}{R} = a_{i1} \ln T + a_{i2}T + \frac{a_{i3}}{2}T^2 + \frac{a_{i4}}{3}T^3 + \frac{a_{i5}}{4}T^4 + a_{i7} \quad (9)$$

Table 3 contains values of the coefficients a_{ij} for CO_2 , H_2O , CO , H_2 , O_2 and N_2 from the NASA program in the range 1000 to 5000 K, which are suitable for calculating the properties of the burned gases.

The thermodynamic properties of the burned mixture, that is, specific heat at constant pressure ($c_{p,b}$), enthalpy (h_b), entropy (s_b), and ratio of specific heat (γ_b), can be obtained using

Table 2 Burned gas composition for fuel-lean conditions under 1700 K.

Component	n_i , moles/mole O_2 reactant
CO_2	$\frac{\zeta\varepsilon}{\lambda}$
H_2O	$\frac{2(1-\varepsilon)\zeta}{\lambda}$
CO	0
H_2	0
O_2	$1 - \frac{\zeta}{\lambda}$
N_2	$\left(1 - \frac{\varepsilon Z}{2}\right)\zeta\psi$
Summation	$n_b = \frac{(1-\varepsilon)\zeta}{\lambda} + 1 + \left(1 - \frac{\varepsilon Z}{2}\right)\zeta\psi$

Equations 10 to 13, with the moles of each species per mole O_2 , n_i , determined from Table 2 and the mass of mixture per mole O_2 , m_b , determined from Equation 5:

$$c_{p,b} = \frac{1}{m_b} \sum_i n_i \tilde{c}_{p,i} \quad (10)$$

$$h_b = \frac{1}{m_b} \sum_i n_i \tilde{h}_i \quad (11)$$

$$s_b = \frac{1}{m_b} \left\{ \sum_i n_i \left[\tilde{s}_i - \tilde{R} \ln \left(\frac{n_i}{n_b} \right) \right] - n_b \tilde{R} \ln(p) \right\} \quad (12)$$

$$\gamma_b = \frac{c_{p,b}}{c_{v,b}} = \frac{c_{p,b}}{c_{p,b} - R_b} \quad (13)$$

where, p is in atmosphere, $R_b = \frac{\tilde{R}}{M_b}$ is the gas

constant for the burned gas and \tilde{R} is the universal gas constant of 8.3143 J/mol·K.

RESULTS AND DISCUSSION

Effects of relative air/fuel ratio on burned gas composition

The calculated gas compositions from the fuel-lean combustion in an engine without exhaust gas recirculation (EGR) are shown in Figure 1 for carbon dioxide (CO_2), water vapor (H_2O), and oxygen (O_2). Nitrogen (N_2) has not been illustrated, as it is comparatively inert and its

fraction was changed insignificantly. Each component, in the form of the mole fraction, was plotted against relative air/fuel ratio (λ) in the range ($1.0 < \lambda < 2.0$), representing real operating load conditions for a diesel engine. The higher the values of the relative air/fuel ratios, the lower the engine loads.

Figure 1 shows the reduction of CO_2 in the burned gases of the RME-air mixture when the relative air/fuel ratio increased. This occurred because the combustion consumed less fuel and emitted less CO_2 at high relative air/fuel ratios. Figure 1 also shows that the H_2O decreased and the O_2 increased in the burned gases, when the relative air/fuel ratio increased. The reduction in the water vapor fraction corresponded to decreased

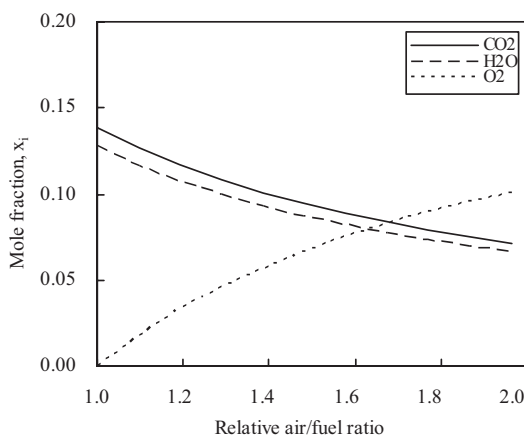


Figure 1 Burned gas composition of rapeseed methyl ester-air mixture as a function of relative air/fuel ratios under fuel-lean conditions.

Table 3 Coefficients for species thermodynamic properties in temperature range 1000 to 5000 K.

Species	a_{i1}	a_{i2}	a_{i3}	a_{i4}	a_{i5}	a_{i6}	a_{i7}
CO_2	0.44608 e+1	0.30982 e-2	-0.12393 e-5	0.22741 e-9	-0.15526 e-13	-0.48961 e+5	-0.98636 e+0
H_2O	0.27168 e+1	0.29451 e-2	-0.80224 e-6	0.10227 e-9	-0.48472 e-14	-0.29906 e+5	0.66306 e+1
CO	0.29841 e+1	0.14891 e-2	-0.57900 e-6	0.10365 e-9	-0.69354 e-14	-0.14245 e+5	0.63479 e+1
H_2	0.31002 e+1	0.51119 e-3	0.52644 e-7	-0.34910 e-10	0.36945 e-14	-0.87738 e+3	-0.19629 e+1
O_2	0.36220 e+1	0.73618 e-3	-0.19652 e-6	0.36202 e-10	-0.28946 e-14	-0.12020 e+4	0.36151 e+1
N_2	0.28963 e+1	0.15155 e-2	-0.57235 e-6	0.99807 e-10	-0.65224 e-14	-0.90586 e+3	0.61615 e+1

Source: Gordon and McBride (1971)

CO₂ emissions, while the increased amount of excess oxygen left in the exhaust gas was attributed to the fuel-lean combustion conditions.

The mass and mole number (per mole of O₂ reactant) of the burned gas of the RME-air mixture are shown in Figure 2. They decreased consistently with the increasing relative air/fuel ratios. These trends were straightforward as seen from Equation 5. However, the reductions led to an insignificant reduction in the molecular weight of the burned gas depicted in Figure 3 (only a 0.15% reduction over the range of relative air/fuel ratios presented).

Effects of relative air/fuel ratio on the thermodynamic properties of the burned gas

The thermodynamic properties of the RME-air burned gas are shown in Figures 4 to 7 for specific heat at constant pressure ($c_{p,b}$), the ratio of specific heats (γ_b), enthalpy (h_b) and entropy (s_b) respectively. The values are plotted against the relative air/fuel ratio (λ) in the fuel-lean burning range ($1.0 < \lambda < 2.0$).

Fundamentally, specific heat, enthalpy and entropy varied with temperature, while the ratio of specific heat did not. As described earlier, the increased relative air/fuel ratios resulted in

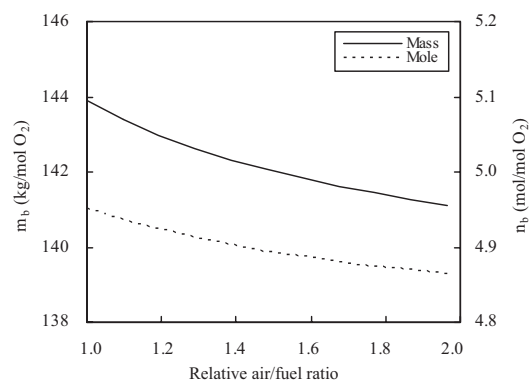


Figure 2 Mass and mole of the burned gas of rapeseed methyl ester-air mixture as a function of relative air/fuel ratios under fuel-lean conditions.

increased excess oxygen and reduced carbon dioxide and water vapor portions in the burned gas. This produced a drastic reduction in the specific heat of the mixture (Figure 4), caused by the higher specific heat portions of CO₂ and H₂O compared to that of O₂. However, the specific heat ratio, based on Equation 13 showed a slight increase with increasing relative air/fuel ratio (Figure 5). At the same temperature, the enthalpy of the burned gas (Figure 6) increased with the relative air/fuel ratio, but the entropy showed the reverse effect (Figure 7). This was mainly due to the variation in the burned gas fractions at various mixing compositions. All these trends in the calculated values were in agreement with the data presented in Heywood (1988), but were dissimilar in quantity due to use of different fuels (C_nH_{2n}) and burning conditions (pressure and fuel-air mix).

The thermodynamic properties (specific heat and specific heat ratio) of the RME-air burned gas at 1500 K are also depicted in Figure 8, in comparison with those of ultra-low sulfur diesel (USLD)-air burned gas. The trend of those values, as well as other temperatures (not illustrated) is consistent in the range 1000 to 5000 K. Greater specific heat values and the subsequent smaller

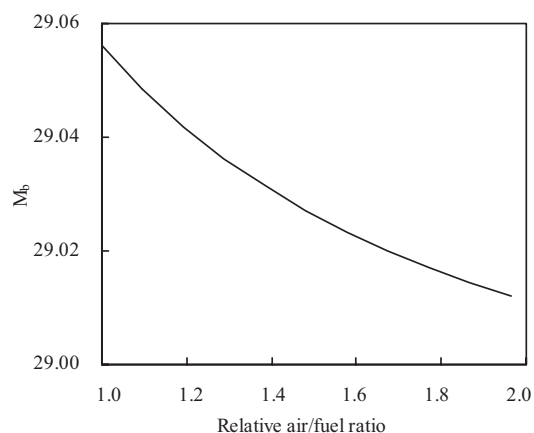


Figure 3 Molecular weight of the burned gas of rapeseed methyl ester-air mixture as a function of relative air/fuel ratios under fuel-lean conditions.

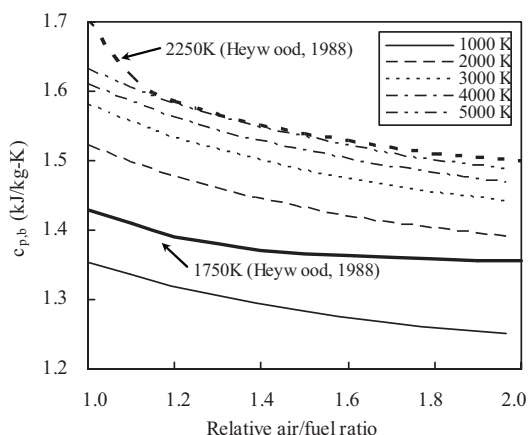


Figure 4 Specific heat at constant pressure of the equilibrium burned gas of rapeseed methyl ester-air mixture as a function of relative air/fuel ratios under fuel-lean conditions at $T = 1000, 2000, 3000, 4000$ and 5000 K. Data comparison with isooctane-air combustion from Heywood (1988).

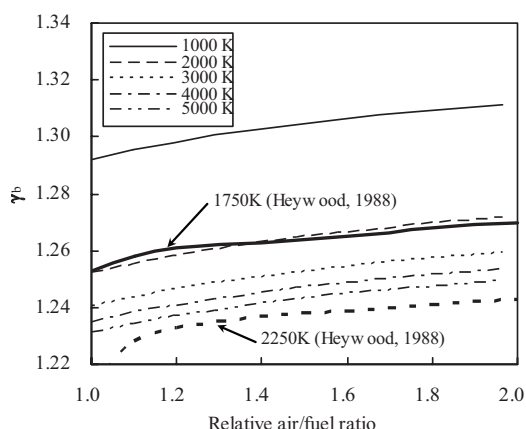


Figure 5 Ratio of specific heats of the equilibrium burned gas of rapeseed methyl ester-air mixture as a function of relative air/fuel ratios under fuel-lean conditions at $T = 1000, 2000, 3000, 4000$ and 5000 K. Data comparison with isooctane-air combustion from Heywood (1988).

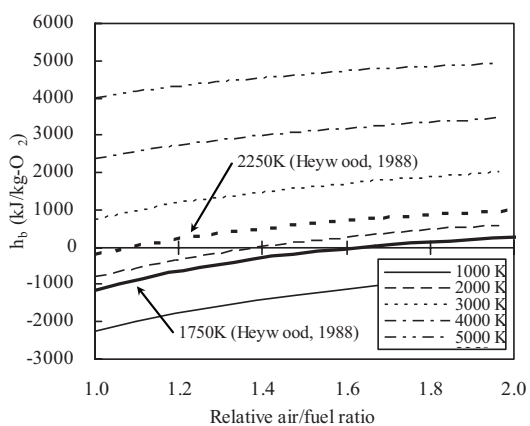


Figure 6 Standard state enthalpy of the equilibrium burned gas of rapeseed methyl ester-air mixture as a function of relative air/fuel ratios under fuel-lean conditions at $T = 1000, 2000, 3000, 4000$ and 5000 K. Data comparison with isooctane-air combustion from Heywood (1988).

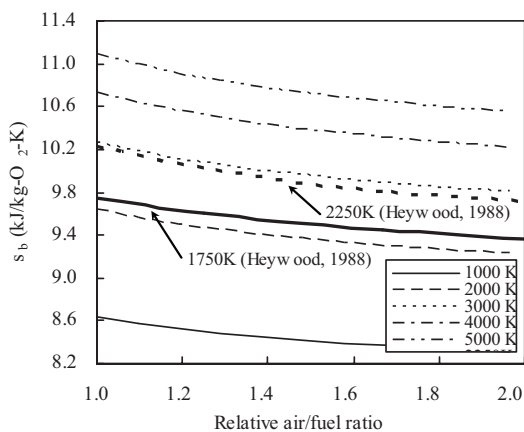


Figure 7 Standard state entropy of the equilibrium burned gas of rapeseed methyl ester-air mixture as a function of relative air/fuel ratios under fuel-lean conditions at $T = 1000, 2000, 3000, 4000$ and 5000 K and 1 atmosphere of pressure. Data comparison with isooctane-air combustion from Heywood (1988).

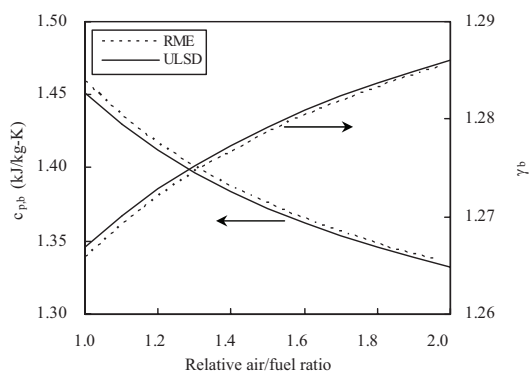


Figure 8 Specific heat at constant pressure and ratio of specific heats comparison between rapeseed methyl ester-air and ultra low sulfur diesel-air burned mixture at 1500 K temperature.

specific heat ratio of the RME-air burned gas were observed over the range of relative air/fuel ratios, compared to those of ULSD-air burned gas. This was because the amount of carbon dioxide and water vapor content in the burned gas, with their higher specific heats, were higher in the RME-air mixture, as confirmed by the experimental results of Chuepeng *et al.* (2008b) (see Table 4 for comparison). This also agreed with theoretical combustion calculations, as the larger carbon and oxygen atoms, as well as the lower calorific value of the fuel reactant (see Table 1 for properties) tended to yield higher amounts of carbon dioxide and water vapor in the burned gas.

In such models, based on the thermo-chemistry of fuel and air combustion, it is necessary to know the thermodynamic properties of the gas mixture, such as the properties presented. In addition, these properties are also crucial for

explaining how that mixture will behave across the engine operating range (load or relative air/fuel ratio). Therefore, the data obtained can be considered as the basis for further calculations on the thermo-chemistry of RME-air combustion analysis. Furthermore, this process may be applied to other kinds of oxygenated fuels, as well as to biodiesel fuels.

CONCLUSION

The thermodynamic properties of the burned gas in mixtures of rapeseed methyl ester-air under fuel-lean conditions were presented. The relative air/fuel ratio that affected the burned gas composition was predicted using a simple analytical model developed for oxygenated fuel. This resulted in the presentation of differences in the thermodynamic properties over a range of relative air/fuel ratios. The calculated specific heat, enthalpy and entropy increased with temperature, but the specific heat ratio did not. At the same temperature, an increase in the relative air/fuel ratio resulted in a reduction in the specific heat and the entropy of the burned gas, while the specific heat ratio and the enthalpy increased. The specific heat and the specific heat ratio of the RME-air burned gas were compared with those of ULSD-air burned gas. The trends of the calculated values were consistent with data presented in the literature.

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Table 4 Experimental burned gas data. *

Fuel	λ	O ₂ (Vol.%)	CO ₂ (Vol.%)	H ₂ O** (Vol.%)
ULSD	3.83	16.74	2.8	2.55
RME	2.93	15.60	3.5	3.16

Source: Chuepeng *et al.* (2008b)

* Burned gas from a V6 2.7L diesel engine at 10% load at 1500 rpm.

** H₂O estimated from the exhaust gas composition.

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