

Effect of the Degree of Unsaturation of Biodiesel Fuels on NOx and Particulate Emissions

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ABSTRACT

The effect of the biodiesel feedstock on the engine performance and emissions is expected to become increasingly important as the emissions regulations become more stringent (Euro 5). This work aims to study the effect of the degree of unsaturation of a biodiesel fuel (which is a characteristic of the original oil), this being quantified by the iodine number, on the pollutant emissions and combustion timing. Four biodiesel fuels with iodine numbers ranging from 90 to 125 were tested pure and blended (30% and 70% biodiesel content, volume basis) with a diesel reference fuel, which was tested too, in a four-cylinder, 2.2 litre, turbocharged, direct injection diesel engine. The operation modes were selected to be representative of the New European Driving Cycle. In general, pure biodiesel fuels, compared to the reference fuel, resulted in sharp reductions in particle mass and opacity (60-70%) and in a slight increase in both fuel consumption (around 15% in mass) and NOx emissions (9%). Additionally, the degree of unsaturation of biodiesel fuels was found to have significant effects on these emissions. As the biodiesel fuel became more unsaturated, NOx emissions increased by 10% and particle mass emissions decreased by 20%. Regarding particle size distributions, unsaturated biodiesel fuels showed a smaller mean diameter. Finally, the effect of iodine number on biodiesel combustion timing was also determined. Experimental results showed a delayed start of ignition but a higher heat release rate as biodiesel iodine number was increased.

INTRODUCTION

Nowadays, the use of biodiesel is being promoted by the European Union to partly replace fossil diesel fuel consumption, and its market share is predicted to significantly increase in the next few years. This increase is necessary to meet the target established by the European Parliament for 2010, which is a biofuel market share of 5.75% (energy basis). Countries like Spain have

just approved biodiesel fuel consumption targets, which will be mandatory in 2009.

Biodiesel is produced from a wide variety of vegetable oils and animal fats. Although some projects or studies using non-conventional feedstock are reported [1, 2], the most common feedstock are soybean, rapeseed and waste cooking oil [3, 4, 5]. Others feedstock are typical in different regions, such as sunflower oil in the Mediterranean area and palm oil in South America and Malaysia. This feedstock could have some effect on engine performance and emissions [6, 7].

Although both prices and availability of different oils in the market are by far the most decisive factors affecting the choice of the oil feedstock by biodiesel manufacturers, the effect of the biodiesel feedstock (more or less unsaturated) on the engine performance and emissions is expected to become increasingly important as the emissions regulations become more stringent (Euro 5).

Previous literature has shown the effect of the degree of unsaturation of biodiesel on its physical and chemical properties. It is accepted that increasing unsaturation improves cold flow properties [8, 9], worsens oxidation stability [8, 10], increases density [6] and reduces viscosity and cetane number [11, 12]. Other effects such as polymerization and deposit formation are often attributed to unsaturation as well [13]. However, much less is published about the effect of the degree of unsaturation of biodiesel fuels and of its derived properties on the engine performance and emissions. All these effects should be taken into account in the discussions currently taking place in the Biodiesel Task Force (subgroup of the Working Group 24 of the European Normalization Committee -CEN) about the possibility of extending the maximum iodine number beyond 120 (as established in the norm EN-14214) in order to open the potential production of biodiesel from new oil feedstock.

Among these emissions, nitrogen oxides (NO_x) are probably the most studied ones in relation to the biodiesel feedstock. In general, authors report that the highest NO_x emissions correspond to the most unsaturated biodiesel fuels, as a consequence of the effect of their physical properties on the injection process or their lower cetane number [8, 14, 15]. Such effects are more appreciated in engines with pump-line-nozzle injection systems than in modern common-rail injection systems, although the latter showed the same trend in recent works [16]. However, the effect of biodiesel feedstock on exhaust gas opacity and particulate emissions is much less known. There is no consensus about whether or not the degree of unsaturation of biodiesel fuels affects smoke opacity and/or particulate emissions [7]. Although a few authors found a slight dependency [17], there are also others who found no effect of the biodiesel feedstock on exhaust opacity and particulate emissions [18].

The present work further investigates the effect of the degree of unsaturation of the biodiesel fuel on the performance and emissions of the engine. The effect on particle size distributions has also been analysed, and constitutes a considerable novelty compared to other published studies.

EXPERIMENTAL FACILITIES

The whole experimental installation is presented in Figure 1, and it has been extensively described in previous works [19, 20]. The engine tested was a 4-cylinder, 4-stroke, turbocharged, intercooled, direct injection diesel engine, typical of those used in European cars. Its specifications are given in Table 1. The engine was connected to an asynchronous electric brake Schenck Dynas III. The engine EGR valve was driven by a stepper motor, which allows to set, for each operation mode, the same EGR ratio regardless of the fuel tested.

Particulate matter was collected using a partial flow dilution minitunnel (Nova Microtroll) equipped with mass flow controllers to achieve the dilution ratio specified by the operator. This minitunnel, as well as the filters, electronic balance and climatic chamber used in the present work is described in [19].

Particle size distributions were determined using a scanning mobility particle sizer (SMPS) TSI 3080. Particles were sampled from the dilution minitunnel, and the largest ones were removed by a 610 nm impactor. These particles are counted in a condensation particle counter (CPC). A correction for particles with multiple charges was automatically applied by the control software.

An opacimeter AVL 439 was used to measure the smoke opacity, which was correlated with FSN (filter smoke number) using the equation proposed in reference [21]. FSN was used to estimate the soot emissions through the recommended correlation by Christian [22].

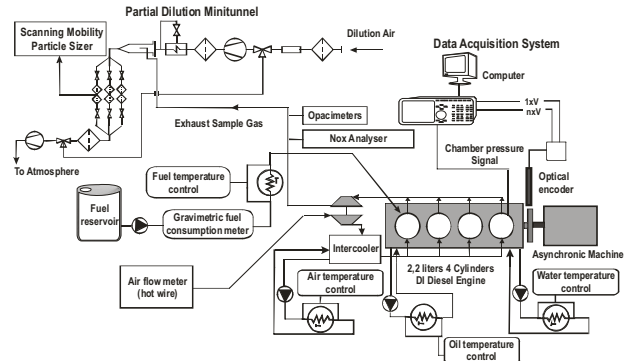


Figure 1 Scheme of experimental facilities

Table 1 Engine characteristics

Fuel injection system	VP44 pump electronically controlled
Injection pressure	200 bar at idle (750 min ⁻¹) 1100 bar at full load (at 2000 min ⁻¹)
Max. rated power	85.2 kW (at 4000 min ⁻¹)
Max. rated torque	237.4 Nm (at 2000 min ⁻¹)
Cylinders arrangement	4, in line
Bore (mm)	86.5
Stroke (mm)	94
Swept volume (l)	2.2
Compression ratio	18:1

The NO_x emissions were measured by a quimioluminescence analyzer Environnement Topaze. A zero dimensional thermodynamic model [23, 24] was used to determine the main parameters of the combustion process such as the start of combustion (SOC) and the heat release law (HRL), from the cylinder pressure signal determined by averaging 20 pressure cycles obtained with a piezo-electric transducer Kistler Z17090.

FUELS

A conventional low sulfur diesel fuel (reference fuel, REF) and four biodiesel fuels (B1 to B4) from different feedstock were tested. In all cases the methyl ester content was above 96.5 % as required in the norm EN-14214. Biodiesel fuels were selected to have different values of iodine number, from 90 to 125. The iodine number increases in proportion to the mean number of molecular double bonds. Besides pure fuels, 30% v/v blends (B1_30, B2_30, B3_30, B4_30) and 70% v/v blends (B1_70, B2_70, B3_70, B4_70) were also tested.

The reference fuel is a typical diesel fuel similar to those available in Spanish petrol stations. It was supplied by Repsol-YPF. Three of the biodiesel fuels (B1 to B3) were produced from waste cooking oils with different composition, and consequently, their iodine values were also different. The estimated composition of the most saturated one (B1: iodine number 91) showed a high percentage of olive oil (saturated oil) while the most unsaturated one (B3: iodine number 115) showed a high content of sunflower oil (unsaturated oil). The last biodiesel fuel, B4, was even more unsaturated than the three waste cooking oil ones (iodine number 125), and it was produced from neat sunflower oil. The composition

and main properties of the pure fuels are presented in Table 2.

Table 2 Fuel properties

	Ref	B1	B2	B3	B4
ρ (kg/m ³) ^a	833.5	885	883.5	880.5	886
ν (cSt) ^b	2.79	5.92	4.92	4.58	4.40
LHV (MJ/kg) ^c	42.84	36.92	36.87	37.01	37.08
% C (in weight)	85.23 ^e	76.90 ^d	76.91 ^d	77.07 ^d	77.15 ^d
% H (in weight)	13.92 ^e	12.19 ^d	12.13 ^d	12.03 ^d	11.96 ^d
% O (in weight)	0.74 ^e	10.91 ^d	10.96 ^d	10.90 ^d	10.89 ^d
ppm S (in weight)	267 ^e	~ 0	~ 0	~ 0	~ 0
MW ^f	206.9 ^g	293.08 ^d	291.86 ^d	293.41 ^d	293.61 ^d
AFR _{st} ^h	14.60	12.56 ^d	12.54 ^d	12.52 ^d	12.51 ^d
Cetane Number	52.9 ^e	58.5 ⁱ	56.5 ⁱ	53.5 ⁱ	51.3 ⁱ
IN (mg/g sample) ^j	~ 0	91.34 ^d	98.37 ^d	114.95 ^d	125.43 ^d

^a Density, measured at 15°C; ^b Kinematic viscosity, measured at 40°C; ^c Low heating value; ^d Obtained from speciation into methyl esters by chromatography; ^e Measured by supplier; ^f Molecular weight; ^g Calculated using the Aspen-Advisor software; ^h Stoichiometric air-fuel ratio; ⁱ Estimated from cetane numbers of individual methyl esters [25]; ^j Iodine number

EXPERIMENTAL METHOD

Five engine operating modes were selected among the collection of steady operation modes that reproduce the New European Driving Cycle that the vehicles equipped with this engine must follow, as fully described in [19]. Each mode was characterized by its engine speed (n), effective torque (Me), brake mean effective pressure ($bmeP$) and EGR ratio (EGR). These values are listed in Table 3.

After each fuel was tested, lines were drained prior to filling them with the following fuel. Also, the sample line of the dilution minitunnel was cleaned in order to eliminate deposits and hydrocarbons from the previous test. After starting the engine, it was warmed up with the new fuel for at least one hour to purge any of the remaining previously tested fuel from the engine fuel system, and to reach the appropriate thermal conditions.

Each fuel was tested once, except the reference diesel fuel, which was tested four times. Performance and emissions of the engine showed no shift with time. Results presented for the reference fuel are the average of the four tests. Error bars indicating 90% confidence level are also provided.

As in other works presented by this research group [19, 26], the duration of each operation mode was chosen so that three different particulate filters could each accumulate a particulate sample of at least 1.3 mg and six particle size distributions could be taken with the SMPS.

Table 3. Operation modes.

Operation mode	Effective Torque (Nm)	Speed (rpm)	$bmeP$ (bar)	EGR ratio
C'	26	1410	1.48	38
F	46	1526	2.66	32
G	55	1661	3.17	28
G'	77	1743	4.44	20
H	110	1853	6.36	11

RESULTS AND DISCUSSION

Results obtained for a) brake specific fuel consumption, b) thermal efficiency, c) smoke opacity, d) particulate matter mass, e) size of particles, f) NOx emission and g) combustion timing are presented in this section. In all the cases the information is shown in a first figure and then reorganised in two different figures to study the effect of the biodiesel content and its feedstock. In this analysis, a linear equation (1) was fitted for each biodiesel feedstock.

$$y = a \cdot z_b \quad (1)$$

where:

y : any variable related to engine performance or emissions.

a : coefficient to be adjusted.

z_b : volumetric fraction of biodiesel in the fuel, $0 \leq z_b \leq 1$. ($z_b = 0$ for the reference diesel fuel and $z_b = 1$ for any of the four pure biodiesel fuels tested).

A new parameter, named as EV_{B100} (expected variation for pure biodiesel fuel), was calculated as the value obtained from the linear equations above described in the cases of pure biodiesel fuels ($z_b = 1$). It is easily observed that this parameter coincides with coefficient a in equation (1).

a) Brake specific fuel consumption ($bsfc$)

An increasing trend in brake specific fuel consumption $bsfc$ was observed when decreasing the load (Figure 2), because of the higher contribution of the mechanical losses to the total power output at low load. In the same figure, increases in $bsfc$ when biodiesel content in the fuel was increased are shown. This was due to the methodology followed (equal speed and effective torque regardless of the fuel) and the lower energy content of biodiesel fuels compared to that of diesel fuels [7, 8], Table 2.

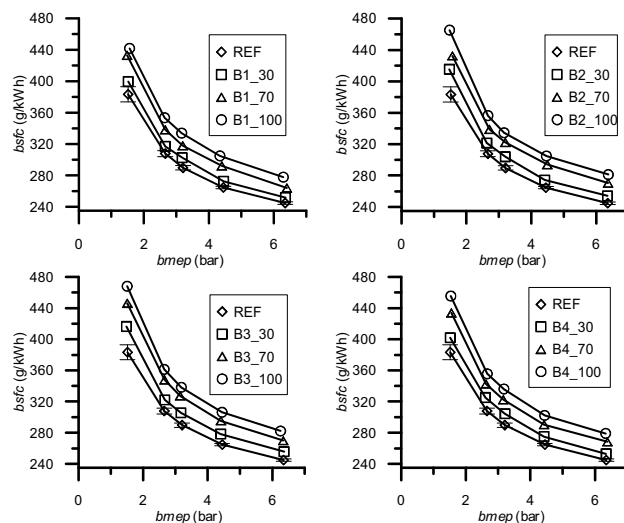


Figure 2 Brake specific fuel consumption