

Potential of Cellulose-Derived Biofuels for Soot Free Diesel Combustion

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ABSTRACT

Today's biofuels require large amounts of energy in the production process for the conversion from biomass into fuels with conventional properties. To reduce the amounts of energy needed, future fuels derived from biomass will have a molecular structure which is more similar to the respective feedstock.

Butyl levulinate can be gained easily from levulinic acid which is produced by acid hydrolysis of cellulose. Thus, the Institute for Combustion Engines at RWTH Aachen University carried out a fuel investigation program to explore the potential of this biofuel compound, as a candidate for future compression ignition engines to reduce engine-out emissions while maintaining engine efficiency and an acceptable noise level.

Previous investigations identified most desirable fuel properties like a reduced cetane number, an increased amount of oxygen content and a low boiling temperature for compression ignition engine conditions. Depending on the chain length, fuel compounds vary in cetane number and boiling temperature. Therefore different blends of butyl levulinate and n-tetradecane, a long-chain alkane, were investigated in this study.

To gain knowledge about the combustion process with biofuels, experiments on a single cylinder diesel research engine were performed. Engine results with blends of butyl levulinate and n-tetradecane are compared to regular diesel fuel and rapeseed oil methyl ester (B100) with respect to all regulated emissions.

With the use of butyl levulinate the soot emissions can be significantly reduced up to 100 % depending on the load point. Furthermore, the experimental results indicate an

additional potential for future diesel combustion concepts such as HCCI with respect to soot and NO_x emissions in the part load range when using oxygenated fuels derived from biomass. Overall, the low particulate matter emissions provide justification for the consideration of butyl levulinate as a candidate for a fuel or fuel compound in future diesel engines. As a drawback malfunctions in various components of the fuel system like fuel hoses and fuel sealings can occur with the standard engine equipment.

INTRODUCTION

The finite nature and instability of fossil fuel supply in combination with the carbon dioxide issue has led to an increasing and enduring investigation demand in the field of alternative and regenerative fuels. Renewable biological fuels have the potential to maintain a constant level of greenhouse gas emissions by not adding any additional fossil carbon to the atmosphere. These fuels can be tailored to the demands of modern engines taking into account the catalytic production processes. With the establishment of these fuels as an additional degree of freedom, a simultaneous optimisation of the engine and fuel is possible.

Through the application of optimised synthesis processes, the Cluster of Excellence "Tailor-Made Fuels from Biomass" (TMFB) adopts an interdisciplinary approach towards research of new, biomass-based synthetic fuels. With regard to modern combustion technologies, this approach was chosen in order to verify their potential, while simultaneously reducing the dependence on fossil fuels. The long-term goal is to determine the optimal combination of fuel components and their production processes, which is based on renewable raw materials and new combustion processes.

Such a tailor-made fuel is a well-defined blend of distinct molecular components with optimised physiochemical properties for future combustion systems, which can be

produced by sustainable and economical production processes.

In the search for new fuels to reduce engine emissions generally and particularly soot emissions several investigations have been recently published [1, 2]. All of them show a positive impact on particulate matter emissions with an increasing amount of fuel based oxygen in the combustion chamber. Soot formation can be reduced linearly with increasing oxygen content [3, 4]. The potential of oxygenated fuels has been previously explored on the example of 1-decanol, a long-chain alcohol. A reduction by up to 90% in particulate matter emissions in comparison to conventional diesel fuel could be identified [5].

Within the Cluster of Excellence TMFB, investigations have recently focused on butyl levulinate (C₉H₁₆O₃) as a candidate for future fuel compounds. This oxygenated hydrocarbon can be easily produced through acid catalysis of cellulose. Despite its very low cetane number, fuel properties enable an utilization as a fuel in a compression-ignited engine exclusively or as part of a blend. Therefore, different blends of butyl levulinate and n-tetradecane have been investigated (butyl levulinate vol-% varying from 60 % to 80 %). The potential of such blends to reduce soot emission will be particularly analyzed in this paper and will be compared with baseline diesel fuel and pure rapeseed oil methyl ester, a first generation biofuel.

TEST SET UP

The engine used in this study was optimized to achieve best combustion behavior for lowest engine-out emissions and highest fuel efficiency. The hardware enhancements include high fuel injection pressures for improved mixture formation, high EGR levels and intensified charge air and EGR cooling to lower the temperature in the combustion chamber for a cooler combustion. Details of the test engine are listed in Table 1. Further information about the research single cylinder test engine are available in different publications [6, 7].

The research engine is capable of achieving Euro 6 emissions performance by means of a lower compression ratio, a higher maximum cylinder peak pressure, a maximum rail pressure of 2000 bar and intensified EGR cooling. These engine enhancements are expected to be commonly used on advanced diesel engines in the near future. As such, the testing capabilities on the engine exceed those of today's production engines.

ENGINE OPERATION - Engine tests for the fuel investigations were carried out in four part-load points, as shown in Figure 1.

Table 1. Hardware specifications for the research engine

	Units	Engine
Emission Status	-	Euro 6
Displacement	cm ³	390
Stroke	mm	88.3
Bore diameter	mm	75
Compression ratio	-	15
Valves per cylinder	-	4
Maximum peak pressure	bar	220
Fuel injection system specifications:	-	Bosch Piezo Common Rail System
Maximum injection pressure	bar	2000
Hydraulic Flow Rate (HFR)	cm ³ /30 s	310
Nozzle hole diameter	μm	109
Number of spray holes	-	8
Spray Cone Angle	°	153
Charging	-	Max. 3.5 bar absolut

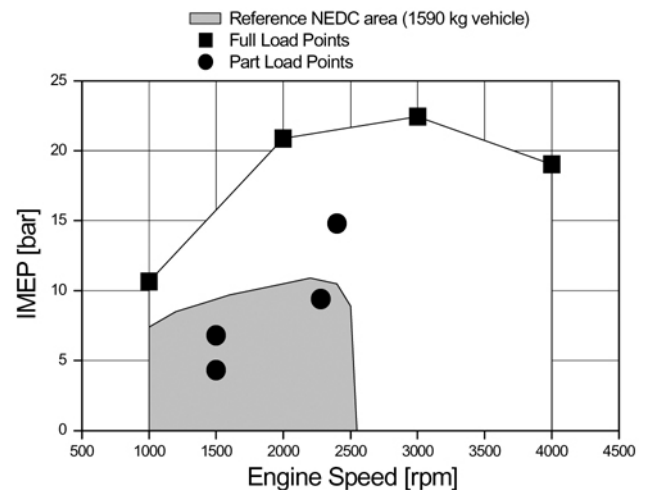


Figure 1. Engine Operating Points

Three of the four part-load points are located within the reference area of the New European Driving Cycle (NEDC) while the highest load point at 2400 rpm and 14.8 bar IMEP is outside this range. Based on the new European Driving Cycle (NEDC) the maximum Indicated Specific NO_x (ISNO_x) and Indicated Specific PM (ISPM) emissions were

Table 4. Engine Calibration of different load points

	Centre of Combustion	Pilot Offset	DOI-Pilot	Rail Pressure	Intake Temp.	Intake Pressure
	°CA BTDC	°CA	μs	bar	K	bar abs
1500 rpm, 4.3 bar IMEP	-6.6 @ 0.5 g/kWh ISNO _x	10	180	720	298	1.07
1500 rpm, 6.8 bar IMEP	-5.8 @ 0.5 g/kWh ISNO _x	11	140	904	303	1.5
2280 rpm, 9.4 bar IMEP	-9.2 @ 0.5 g/kWh ISNO _x	20	120	1399	308	2.29
2400 rpm, 14.8 bar IMEP	-10.8 @ 1.0 g/kWh ISNO _x	28	120	1800	318	2.6

estimated for each part-load point and different Euro emission levels for a 1590 kg vehicle. These targets take into account the expected impact of transient operation and cold start and include an appropriate engineering margin. The estimated NO_x and PM target levels are summarized in [Tables 2](#) and [3](#). Investigations on a chassis dynamometer confirmed these estimated targets

Table 2. Estimated Indicated Specific NO_x (ISNO_x) Targets Required to Meet Different Euro Emissions Limits at the Four part-load Operating Points

ISNO _x [g/kWh]	1500 rpm, 4.3 bar IMEP	1500 rpm, 6.8 bar IMEP	2280 rpm, 9.4 bar IMEP	2400 rpm, 14.8 bar IMEP
Euro 4	0.6	0.6	1.8	2.25
Euro 5	0.4	0.4	1.2	1.50
Euro 6	0.2	0.2	0.6	0.75
50%	0.1	0.1	0.3	0.38

Table 3. Estimated Indicated Specific PM (ISPM) Targets Required to Meet Different Euro Emissions Limits at the Four part-load Operating Points

ISPM [g/kWh]	1500 rpm, 4.3 bar IMEP	1500 rpm, 6.8 bar IMEP	2280 rpm, 9.4 bar IMEP	2400 rpm, 14.8 bar IMEP
Euro 4	0.0030	0.0279	0.0706	0.2129
Euro 5	0.0006	0.0056	0.0141	0.0426
Euro 6	0.0006	0.0056	0.0141	0.0426
50%	0.0006	0.0056	0.0141	0.0426

The experiments were carried out with pilot injection and simulated closed loop combustion control. This was done by adjusting the fuel injection timing on each fuel so that the centre of combustion (CA50) was always located at the same degree crank angle in the engine cycle, chosen to maximise the engine efficiency, regardless of the fuel properties. Fuels with lower cetane number (CN) and longer ignition delay require earlier fuel injection timing to maintain the centre of combustion at the required crank angle position. Therefore the begin of injection varies between 5 °CA BTDC at 2400 rpm, 14.8 bar IMEP and 20 °CA BTDC at 1500 rpm and 4.3 bar IMEP. The difference between the energizing time of the injector and the begin of injection was measured to 160 μs. The conditions at the four part-load operating points were chosen with respect to today's achievable ranges and are listed in [Table 4](#). The intake manifold temperature was measured and controlled directly after the intercooler and before the EGR mixing.

<[table 4 here](#)>

FUEL PROPERTIES - The potential of fuels which can be derived easily from biomass was in the focus of interest within this study. It is common knowledge that Cellulose, separated from lignin and hemicellulose, will serve as a starting molecule for the biogenous fuel production. The separation of oxygen from the cellulose requires high amounts of hydrogen and is therefore not desirable from an energetic point of view. In the case of a Fischer-Tropsch fuel (2nd generation biofuel), where the feed stock will be broken up into smallest molecules, a separation is not necessary anymore. Within the cluster of excellence TMFB completely new production routes based on catalytic processes will be developed. These fuels will most likely contain oxygen. One of these fuel candidates is butyl levulinate which belongs to the group of levulinic esters, which can be converted from

levulinic acids by dehydration/hydrogenation or esterification, while levulinic acid is directly produced by acid hydrolysis of cellulose. A molecular structure of butyl levulinate is shown in [Figure 2](#). The low cetane number and high oxygen content of levulinic esters are ideal fuel properties for future premixed charged compression ignition combustion systems. On the other hand the commercial use of levulinic esters is limited due to the high cost of their production.

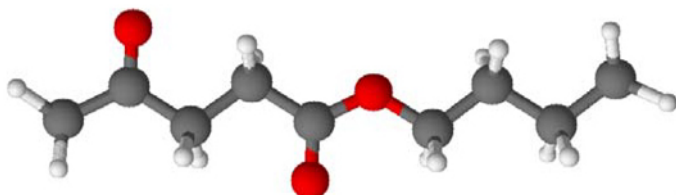


Figure 2. Molecular composition of butyl levulinate

However, due to the similar molecular structure of the fuel compound and the respective feedstock, the production of these esters from cellulosic biomass represents a potential low-cost route. Estimations have shown that levulinic esters can be produced on a large scale at less than 0.50 euro/L [8].

Today, levulinic esters are already known as an additive for Diesel fuel to increase the oxygen content [9]. Butyl levulinate consists of about 30% oxygen. Due to the molecular structure, particularly the small number of c-atoms and the oxygen content of an ester, butyl levulinate has a cetane number below the measurement limit, which is 18 in an ignition quality tester (IQT), the tester used for this study. Thus, cetane number improvers are needed to ignite the fuel in a compression ignition engine. Butyl levulinate can not be used as a pure chemical to substitute the fuel but needs to be blended with fast burning molecules. Therefore n-tetradecane, a long-chain paraffin with a cetane number of 95, was selected. N-tetradecane can not be easily derived from biomass. But its high cetane number provides justification for the consideration as a fuel blend. In future n-tetradecane can be substituted by other high cetane number fuels like GtL or BtL fuel. The most appropriate fuel composition was investigated in a first step.

To gain valuable information about the potential of future fuels from biomass, promising fuel candidates need to be compared to today's available fuels. Therefore, additional investigations were carried out with Fatty Acid Methyl Ester (FAME). The FAME used in this study was a Rapeseed Methyl Ester (RME) that complied with EN 14214 specifications. Rapeseed Methyl Ester is a 100 % first generation biofuel and therefore called B100 in this paper. It consists of saturated as well as unsaturated fatty acids with 12 to 22 carbon atoms. The catalytic reaction is initiated by adding methanol to rapeseed oil. The outcome of this process

is Rapeseed Methyl Ester as well as raw glycerine. Using the difference in density, B100 and glycerine can be separated. B100 is the most common fossil diesel-like substitute in the European Union.

In the following all test results will be compared to available Diesel results which were carried out in a previous study using the same engine calibration [10]. The Diesel fuel meets the requirements of the EN 590 specifications.

The corresponding properties of all investigated fuels are listed below in [Table 5](#).

<table 5 here>

RESULTS

To evaluate the benefit of optimized fuel properties attention naturally focuses on the part-load test conditions that influence performance in the NEDC emissions cycle. For this reason, this section is devoted to an assessment of these part-load conditions.

Engine measurements were taken by varying the EGR rates over a wide range to adjust NO_x emissions for the corresponding Euro-level. All results presented in this paper are illustrated at the Euro 6 NO_x emission limit.

The analysis of butyl levulinate as a candidate for future fuel compounds or even as fuel surrogate was carried out in two steps. At first, different blends of butyl levulinate and n-tetradecane with varying n-tetradecane content from 10vol %-40vol% were investigated with regard to their combustion behavior, particularly the ignition delay time. These burning characteristic investigations allow valid conclusions about the available time for mixture preparation which is getting a more dominant role for future homogeneous or partly homogeneous combustion systems. Using the most appropriate blend, all emissions including noise emissions were measured in all four load points in a second step.

Overall, this approach allows a conclusion about the potential of butyl levulinate to reduce carbon dioxide emissions and to benefit from a cleaner combustion in future combustion systems at the same time.

INVESTIGATION OF COMBUSTION PERFORMANCE USING DIFFERENT FUEL BLENDS - Due to the low cetane number the blending of butyl levulinate with fast igniting fuel compounds is a prerequisite to start the combustion in a compression ignition engine. Additionally, the blending offers opportunities to adjust desired fuel properties like ignition delay time or oxygen content.

Within this research study three different blends were investigated and extensively analyzed. The ignitibility of a

Table 5. Properties of investigated fuels

	Unit	EN590 Diesel	butyl levulinate	B100	n-tetra-decan
Density (15 °C)	kg/m ³	833.2	978.3	883.2	762
Viscosity (40 °C)	mm ² /s	~3.25	2.00	~5.25	2.05
Carbon content	w-%	86.46	62.77	77.12	84.85
Hydrogen content	w-%	13.4	9.36	12.05	15.15
Oxygen content	w-%	0.14	27.87	10.83	0
Aromatics content	mg/kg	24.9	0	0	0
Molecular weight	g/mol	185	172	283	198
Lower Heating value	kJ/kg	42900	27890	37530	44240
Boiling temperature	°C	180-350	232	352	252
Cetane number	-	56.5	< 18	51.8	95

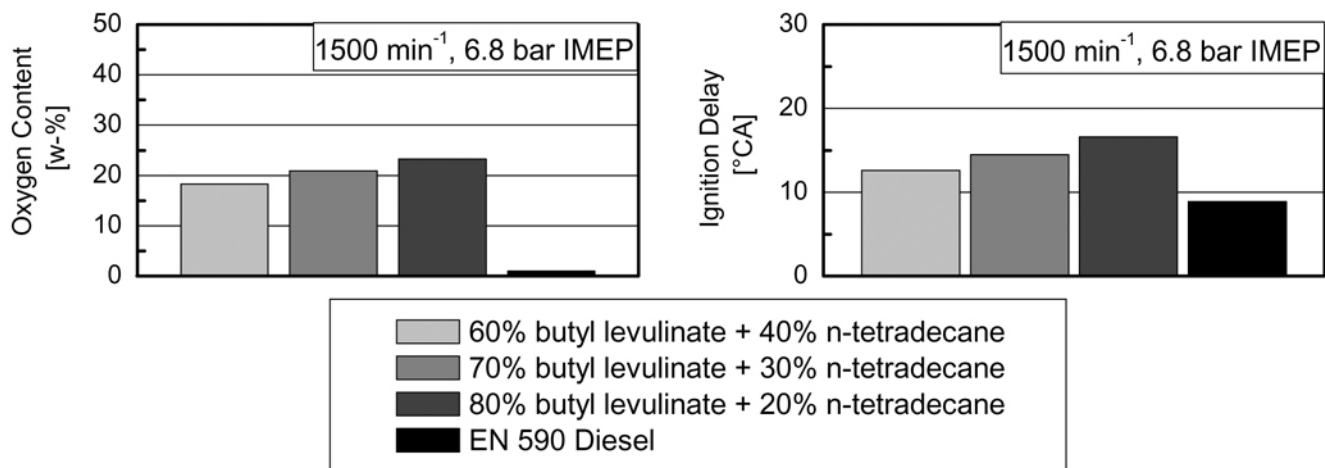


Figure 3. Oxygen content and ignition delay times of investigated fuel blends

air/fuel mixture is highly depending on the intake air temperature and the boost pressure. The combustion chamber temperature needs to be high enough to start the combustion. Thus, lower part load points are more critical concerning the general ability to start the combustion. Therefore, the investigations of different fuel blends were carried out with pilot injection at a low part load point of 1500 rpm and 6.8 bar IMEP. To maintain equal testing settings and to assure valid comparability conditions all measurements were carried out at a constant ISNOx level. Furthermore the centre of combustion was kept constant.

Figure 3 shows the oxygen content and the measured ignition delay time for the three investigated fuel blends compared to standard diesel fuel. The content of the ignition improver n-tetradecane was varied for these investigations from 20- vol % to 40- vol %. The enrichment of only 10- vol % of n-tetradecane was not sufficient to ignite the fuel at lower load conditions.

<figure 3 here>