

Isobutanol as Both Low Reactivity and High Reactivity Fuels with Addition of Di-Tert Butyl Peroxide (DTBP) in RCCI Combustion

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ABSTRACT

Engine experiments and multi-dimensional modeling were used to explore the effects of isobutanol as both the high and low reactivity fuels in Reactivity Controlled Compression Ignition (RCCI) Combustion. Three fuel combinations were examined; EEE/diesel, isobutanol/diesel, and isobutanol/isobutanol+DTBP (di-tert butyl peroxide). In order to assess the relative performance of the fuel combinations of interest under RCCI operation, the engine was operated under conditions representative of typical low temperature combustion (LTC). A net load of 6 bar indicated mean effective pressure (IMEP) was chosen because it provides a wide operable range of equivalence ratios and combustion phasings without excessively high peak pressure rise rates (PPRR). The engine was operated under various intake pressures with global equivalence ratios from 0.28-0.36, and various combustion phasings (defined by 50% mass fraction burned-CA50) from about 1.5 to about 10 deg after top dead center (ATDC). Combustion phasing was varied by scaling the direct-injected fuel quantity appropriately.

Due to isobutanol's high octane number, a significantly larger quantity of direct-injected fuel was required to match combustion phasing, resulting in higher NO_x emissions and decreased gross thermal efficiency.

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INTRODUCTION

Stricter pollutant emission regulations for vehicles have been promulgated by governmental agencies [1] due to concerns over local air pollution. Researchers have sought new and novel combustion strategies to avoid the pollutant emissions issues associated with conventional diesel and spark ignited engines while simultaneously increasing fuel economy [2]. Compression ignition (CI) combustion strategies have been of particular interest because of the potential for high thermodynamic efficiency and low fuel consumption made possible by the use of high compression ratios without knock or detonation [2]. Conventional diesel combustion, however, suffers from high NO_x and PM emissions, and struggles to meet strict emissions regulations without significant and costly exhaust aftertreatment [2]. In an effort to avoid NO_x and PM formation while maintaining the high efficiency of the CI engine, researchers have turned to LTC strategies which avoid in-cylinder regions of high temperature and equivalence ratio, such as homogeneous charge compression ignition (HCCI) and RCCI.

Reactivity Controlled Compression Ignition (RCCI)

Motivated by work of Bessonette et al. [3], who showed that the optimal fuel for HCCI combustion had an octane number between that of diesel and gasoline and that the desired octane number was a

function of engine load, and Inagaki et al. [4], who demonstrated that in-cylinder fuel blending of fuels with different properties could provide good combustion control with low emissions, Kokjohn et al. [5] outlined the RCCI strategy. RCCI involved port injection of a low reactivity fuel and multiple direct injections of a high reactivity fuel to provide both equivalence ratio and reactivity stratification in the cylinder. Work by many researchers [6, 7, 8] and others has demonstrated a wide operable load range for RCCI combustion with low NO_x and PM emissions. A variety of high and low reactivity fuels have been used to demonstrate significant control over the combustion phasing with acceptable peak pressure rise rates. This control is a direct result of the equivalence ratio and reactivity stratification introduced by direct injection of the high reactivity fuel.

Kokjohn et al. [9] showed the importance of both equivalence ratio and reactivity stratification on the combustion event, and demonstrated the large degree of control over the shape and phasing of the combustion event that the RCCI strategy yields. Splitter [10] showed the effect of mixture stratification due to injection timing and number of injections on performance, and demonstrated that the optimal dual fuel strategy occurs between fully premixed, and overly stratified late injection cases. The inherently fuel flexible nature of RCCI allows the strategy to utilize fuels with a very wide range of properties.

Fuel Effects in LTC Strategies

Many studies have been performed to determine the effects of fuels on low temperature combustion strategies including HCCI, and RCCI and to extend the feasible operable range of these strategies.

In order to better understand the effects of fuel composition and properties on RCCI, and to extend the operable limits, significant research has been conducted. Simulations of Nieman et al. [11] using methane as the low reactivity fuel were able to reach 23 bar IMEP while maintaining high efficiency and low emissions with the use of exhaust gas recirculation (EGR). Hanson et al. [12] investigated the use of cetane improved gasoline as a high reactivity fuel for low load operation, and Splitter et al. [6] demonstrated increased gross thermal efficiencies, load extension, and reduced EGR requirements at high loads with the use of E85 instead of gasoline as a low reactivity fuel. It was found that the lower reactivity (higher octane) of E85 led to longer combustion durations, decreased pressure rise rates, reduced EGR requirements, and decreased heat transfer to the piston and cylinder walls. These effects combined to increase the gross thermal efficiency relative to a gasoline/diesel baseline.

Bio and Alternative Fuels

With rising concerns over depletion of fossil fuel and rising global CO₂ emissions, there is increasing interest in renewable, bio-derived fuels which could minimize net CO₂ emissions. Ethanol is the predominant alternative fuel available in the United States, and it is widely distributed in blends with gasoline in percentages from 10-15% by volume (E10-E15) [13]. The United States Environmental Protection Agency (EPA) has mandated renewable standards, which necessitate an increase in biofuel production over the next decade. These standards cap the production of corn-based ethanol at 15 billion gallons, necessitating production of other advanced biofuels [13].

Efforts to increase the maximum allowable percentage of ethanol in gasoline to make better use of ethanol's high octane number have been met with some resistance as older vehicles, lawn equipment, and recreational vehicles such as powerboats are not equipped to deal with the higher alcohol content [14]. However, the EPA has granted a waiver for use of blends up to 15% by volume of ethanol in vehicles built after 2001 [15].

Splitter et al. [16] demonstrated the potential for fuel consumption and tailpipe CO₂ emissions reductions with engine and vehicle optimization using fuels with high alcohol content, such as E30 and iB24 (24% by volume isobutanol with remainder gasoline) in SI engines. With the ever increasing renewable fuel standards and the potential efficiency gains and CO₂ reductions, alcohols and their derivatives stand poised to become a much larger portion of the transportation fuel market. If advanced combustion strategies are to be utilized in production applications, it will be important to understand the impacts of these bio and alternative fuels on advanced combustion applications.

Biofuels in Advanced Combustion Strategies

Significant research has been performed that outlines the benefits of alcohols and biofuels for advanced combustion [6, 8, 12, 17, 18, 19, 20]. The higher octane of alcohol-based fuels is attractive for low temperature combustion strategies that are often limited at higher loads due to high pressure rise rates. The increased reactivity stratification in RCCI with a high octane fuel allows for lower pressure rise rates and a broader heat release rate [6].

Cetane Improvers

Research involving the use of cetane improvers to improve the autoignition of various fuels has been performed by various authors, including Schwab et al. [21] who studied the effects of DTBP and 2-Ethyl-Hexyl Nitrate (EHN) addition on the cetane number of two different diesel fuels and showed that EHN provided a greater increase in cetane number than DTBP, Mack et al. [22] and Eng et al. [23] studied the effect of DTBP blended with fuels in HCCI combustion. Research by Splitter et al. [6, 10, 24] using EHN and DTBP in gasoline, Kaddatz et al. [25] using EHN in E10, Hanson et al. [12] using EHN in gasoline, and Dempsey et al. [20] using DTBP and EHN in ethanol and methanol has shown the ability to operate across a wide load range using a variety of low reactivity fuels and cetane improved low reactivity fuel as the high reactivity fuel source to achieve RCCI combustion. Using a cetane improver could eliminate the need for two separate fuel streams, allowing a low reactivity fuel to be doped with the cetane improver in situ [25].

Butanol as an Advanced Transportation Fuel

Ethanol is the predominant renewable fuel source used in the US today. However, it has a number of properties which make it less desirable as a transportation fuel [13]. As the number of carbons in the alcohol increases, so do the lower heating value (LHV) and the stoichiometric air/fuel ratio (AFR). The primary drawbacks of ethanol in high percentages are its low energy density which leads to increased fuel consumption despite potential efficiency gains, and the low stoichiometric air fuel ratio, requiring alteration of the air/fuel ratio to maintain stoichiometric equivalence ratios in SI engines. The higher alcohols are therefore more attractive from a LHV and AFRstoich basis [13]. A promising alternative alcohol from a transportation perspective is butanol and its isomers. Isobutanol has an octane number in the range of ethanol, and n-butanol has an octane number similar to that of regular pump gasoline. Butanol can also be blended in gasoline in higher quantities than ethanol without modification of the engine system due to its higher stoichiometric air/fuel ratio [14]. Butanol is also very attractive to the recreational marine market, as these engines typically do not employ closed loop air/fuel ratio feedback controls and cannot adapt to high ethanol content in the fuel [14]. Butanol also has the advantage of being hydrophobic, as opposed to ethanol which is fully miscible in water. This means that butanol/gasoline mixtures could be transported via the existing pipeline infrastructure [26].

The motivation of the present study is to develop an understanding of how isobutanol compares to gasoline as a low reactivity fuel in RCCI combustion, and to establish whether or not isobutanol can be used as a

cetane improver is a feasible high reactivity fuel similar to gasoline doped with cetane improver as shown in [6, 10, 12, 25], given the potential benefits of butanol as a transportation fuel.

EXPERIMENTAL SETUP

All engine experiments utilized a heavy-duty 2.44L displacement Caterpillar 3401 Single Cylinder Oil Test Engine (SCOTE). The engine is a single-cylinder variant of the commercially produced Caterpillar 3406E six-cylinder diesel engine, rated at 62kW (83hp) at 1800rev/min. The specific engine geometry can be seen in Table 1.

Table 1. Stock 3401E SCOTE engine geometry

Displacement	2.44L
Bore x Stroke	13.72 x 16.51cm
Connecting Rod Length	21.16cm
Swirl Ratio	0.7
Number of Valves	4
Intake Valve Opening (IVO)	335°CA ATDC Firing
Intake Valve Closing (IVC)	-143°CA ATDC Firing
Exhaust Valve Opening (EVO)	130°CA ATDC Firing
Exhaust Valve Closing (EVC)	-355°CA ATDC Firing

Laboratory Hardware and Setup

The layout of the intake and exhaust air handling systems can be seen in Figure 1.

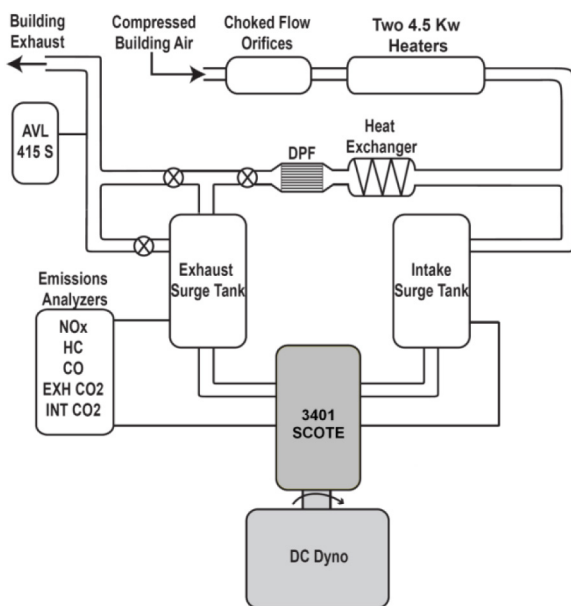


Figure 1. Caterpillar 3401 SCOTE engine.

Intake air flow is metered by a choked-flow orifice rack before passing through two air heaters used to reach the desired intake temperature. The lab simulates a turbocharger by controlling the exhaust and intake surge tank pressures with compressed, dry

building air. The engine is instrumented in various locations with type K thermocouples manufactured by Omega Engineering Inc. Intake temperature was measured at the outlet of the surge tank upstream of the port fuel injector. The exhaust temperature measurement used for the calculation of turbocharger efficiency was measured in the exhaust runner at the point at which the exhaust leaves the cylinder head. The laboratory is also instrumented with various pressure transducers used to determine steady-state operation.

The experiments utilized a 14.88:1 compression ratio piston designed for RCCI combustion. A detailed analysis of the piston design can be found in Splitter et al. [27]. Piston geometry and compression ratio were not the focus of the present research, but a cross sectional view of the RCCI piston profile overlaid over the stock Conventional Diesel Combustion (CDC) piston profile is seen in Figure 2.

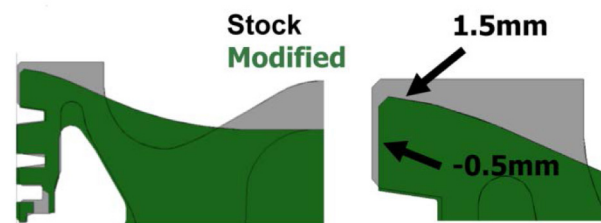


Figure 2. Piston cross section, stock CDC 16.1:1 r_c in grey. Optimized RCCI 14.88:1 r_c in green [27].

Emissions Measurement

Gaseous engine-out emissions were analyzed with a five gas Horiba analyzer bench. Particulates in the lab are measured with an AVL 415s smoke meter, however the smoke meter was not operational during the present experiments, and therefore PM measurements are not reported but are expected to be very low due to the use of an oxygenated fuel in RCCI combustion [6]. The emissions analyzers used in the experiments can be seen in Table 2.

Table 2. Emissions Analyzers

Emission Species	Instrument Manufacturer	Model	Measurement Method
Exhaust NO/NO2	Horiba	CLA-220	Heated Chemiluminescence Detector
Exhaust CO	Horiba	AIA-220	Non-Dispersive Infrared Light Absorption
Exhaust CO2	Horiba	AIA-220	Non-Dispersive Infrared Light Absorption
Exhaust HC	Horiba	FIA-236	Heated Flame Ionization Detector
Intake CO2	Horiba	AIA-220	Non-Dispersive Infrared Light Absorption
Particulate Matter	AVL	415S Smoke Meter	Filter Paper Opacity

RCCI Strategy and Operating Conditions

The standard RCCI strategy involves port injection of a low reactivity fuel to provide a background equivalence ratio in the cylinder, while multiple direct injections of high reactivity fuel between -90 and -25 degrees ATDC create gradients of reactivity and equivalence ratio that provide control over the combustion phasing and decreased peak pressure rise rates compared to HCCI operation [10]. Figure 3 shows the injector locations and injector layout under RCCI operation.

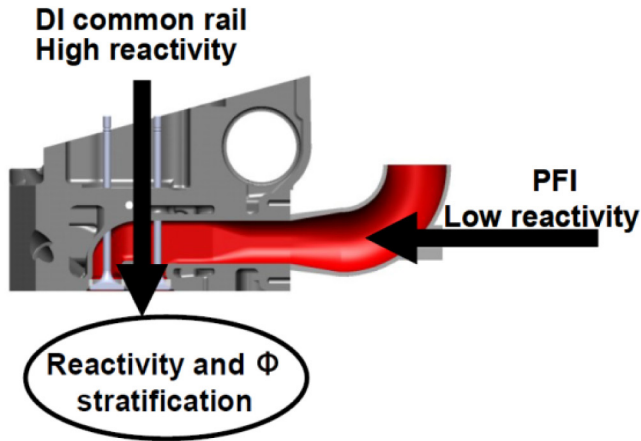


Figure 3. Fuel injection locations and RCCI fueling strategy.

While RCCI is inherently fuel flexible and can operate across wide speed and load ranges by adjusting the ratio of the low and high reactivity fuels [19], the specific fuel properties, including the octane number and heat of vaporization, play a large role in the high reactivity fuel requirements and subsequently on the emissions and performance [20].

Fuels and Fuel Delivery

The port fuel injector specifications can be seen in Table 3. Port fuel was delivered via a lift pump mounted under the fuel tank and the fuel pressure was metered by a regulator, which bypassed excess fuel through a liquid-liquid brazed plate heat exchanger before returning to the fuel tank. Fuel flowrate was measured by an Endress-Hauser Promass 80A01 Coriolis-type flowmeter.

Table 3. Port fuel injector specifications

Manufacturer	RC Engineering
Injector Style	Lucas Peak and Hold
Peak/Hold Current	4.0/1.5Amps
Steady Flow Rate @ 3 bar	750cc/min
Included Spray Angle	15°
Fuel Pressure	3bar

DI fuel was supplied to a Bosch common rail (CR) from a CP3 common rail pump in an external fuel cart. The CR pump was supplied by a lift pump mounted under the fuel tank, and fuel pressure was metered by a regulator, which bypassed excess fuel through a liquid-liquid brazed plate heat exchanger before returning

to the fuel tank. Return fuel from the CR and CR injector passed through a second brazed plate heat exchanger before returning to the CR pump. Fuel flowrate was measured by an Endress-Hauser Promass 80A01 Coriolis-type flowmeter. The common rail injector specifications can be seen in Table 4. The injector body is a commercially available part used in the GM/Fiat 1.9L diesel engine. Various injector tips could be utilized with this injector body, but the current experiments utilized a 6-hole 250 μ m injector tip from an 8.3L high output Cummins QSC-600 marine engine [10].

Table 4. Common rail injector specifications

Manufacturer	Bosch
Style	CRIN-II
Number of Holes	6
Hole Diameter	250 μ m
Included Spray Angle	145°
K Factor	0
Sack Style	Mini-sack
Manufacturer	Bosch

Table 5 lists selected physical and chemical properties of the low reactivity and high reactivity fuels used in this study. Worth noting from the table is the difference in octane number between the EEE and isobutanol. The high octane number of isobutanol could provide potential benefits at high loads with respect to reduced EGR requirements and at conditions where operation would be limited by high peak pressure rise rates [6], but may prove detrimental in terms of efficiency and emissions at lower loads which are the focus of this research.

Table 5. Fuel physical and chemical properties

Fuel	EPA Tier II EEE	isoButanol [26]	Diesel	DTBP [20]
H/C Ratio	1.86	2.5	1.782	2.25
O/C Ratio	0	0.25	0	0.25
Density (g/cm ³)	0.742	0.802	0.856	0.796
LHV [MJ/kg]	43.01	32.96	42.65	33.8
A/F _{stoich}	14.60	11.18	14.46	10.86
RON	96.4	113	N/A	N/A
MON	88.5	94	N/A	N/A
Cetane	N/A	N/A	41.2	N/A

In order to determine the effectiveness of DTBP as an ignition improver, and to determine an adequate addition quantity in isobutanol to achieve a sufficiently reactive fuel mixture, HCCI simulations were performed in KIVA using a reduced PRF mechanism that included ethanol, isobutanol, and DTBP reaction pathways. The mechanism was validated with experimental HCCI

data from Dempsey et al. [20] using a variety of DTBP percentages in methanol and ethanol via a dual port fuel injector intake design. Dempsey first ran primary reference fuels (PRF) isooctane and n-heptane in various ratios and determined CA50. He then matched the combustion phasing of a given cetane improved fuel to a specific ratio of PRF to determine an effective PRF number which corresponds to a given fuel's reactivity and is analogous to octane number. Figure 4 shows the reactivity enhancement of DTBP in methanol, ethanol, and isobutanol.

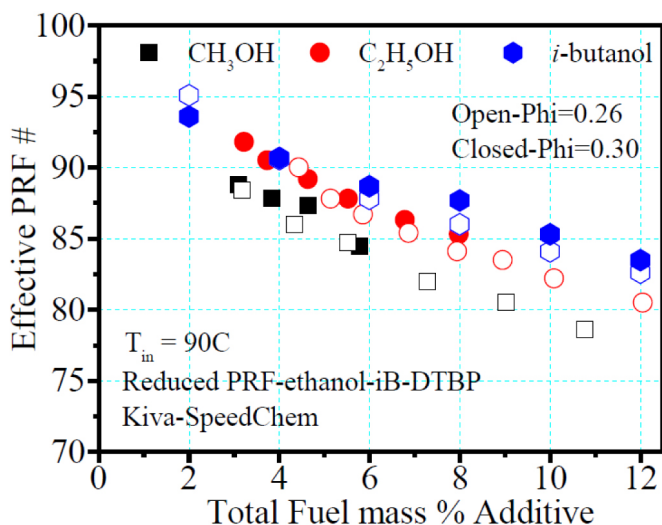


Figure 4. Reactivity enhancement (in terms of effective PRF#) in Methanol, Ethanol, and IsoButanol (GM 1.9 L HCCI combustion simulations). Open symbols represent a global ϕ of 0.26, closed represent ϕ of 0.30.

Based on the simulation results shown in Figure 4 conducted using the chemistry mechanism of Wang et al. [28], a very high DTBP percentage was found to be required to produce a sufficiently reactive mixture for use in RCCI combustion. Accordingly, Isobutanol doped with 20% DTBP by volume (19.9% by mass) was chosen, providing an effective PRF number of about 75-80 (based on extrapolation of the simulation results). The DTBP was splash blended in the fuel tank with the isobutanol, and the system was sufficiently mixed before acquiring data.

Table 6. High reactivity fuel mixture properties

Fuel	Diesel	iBuOH+20%DTBP
H/C Ratio	1.782	2.472
O/C Ratio	0	0.25
Density (g/cm ³)	0.856	0.801
LHV [MJ/kg]	42.65	33.13
A/F _{stoich}	14.46	11.12
Cetane	41.2	N/A

Table 6 shows the fuel properties of the actual high reactivity fuels used in the following sections. A lubricity improver Infineum R655 was splash blended at 500ppm for the iBuOH+20%DTBP fuel.

RESULTS

The three fuel combinations analyzed in the study can be seen in Table 7. EEE/Diesel provides a moderate reactivity gradient between the low reactivity fuel and the high reactivity fuel, IsoButanol/Diesel has a large reactivity gradient, and IsoButanol/IsoButanol+20%DTBP has a relatively small gradient of reactivity between the two fuels.

Table 7. Experimental fuel combinations

Fuel Combination	Low Reactivity Fuel (PFI)	High Reactivity Fuel (DI)
1	EPA Tier II EEE Gasoline	2007 Cert Diesel
2	IsoButanol (99.8% Purity)	2007 Cert Diesel
3	IsoButanol (99.8% Purity)	IsoButanol+20%DTBP

Engine Operating Conditions and Sweeps

In order to assess the relative performance of the fuel combinations of interest the engine was operated under a net load of 6 bar IMEP was chosen this load condition provides a wide operable range of equivalence ratios and combustion phasings without excessively high peak pressure rise rates. The engine operating conditions can be seen in Table 8.

Table 8. Engine operating conditions

Speed (rev/min)	1300
Load (IMEPn, bar)	6±0.1
CA50 (°CA ATDC)	1.5-10 (varied)
Piston Bowl	Bathtub
CR (-)	14.88
Intake Temperature (°C)	40
Intake Pressure (bar)	1.24-1.65 (varied)
Exhaust Pressure (bar)	1.48-1.96 (varied)
Turbocharger η (%)	~50
Low Reactivity Fuel (PFI)	See Table 7
High Reactivity Fuel (DI)	See Table 7

The equivalence ratio was determined by setting the intake surge tank pressure with a simulated turbocharger efficiency of about 50%, as defined by Splitter [10]. The direct injection schedule utilized in the study can be seen in Table 9. The injection schedule remained constant throughout the experiments, but the duration of both direct injections were scaled to reach the required DI quantity to control the combustion phasing, as defined by CA50.