Numerical study of a boosted HCCI engine fueled with n-butanol and isobutanol

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ABSTRACT

Butanol, as a fuel in internal combustion engines, has many advantages over ethanol as biologically derived component of gasoline. The benefits include higher energy density, less water solubility, and better engine compatibility. This study assesses four different chemical kinetic mechanisms for two butanol isomers (n-butanol and isobutanol) in a single-zone engine model and compares the results to experiments conducted on a Homogeneous Charge Compression Ignition (HCCI) engine. The simulations, and experiments, spanned a range of intake pressures (1.0–1.8 bar) and equivalence ratios (0.3–0.4). As was seen in the experiments, the numerical model was able to reflect delayed combustion timings as the intake temperature was decreased. Additionally, the mechanisms were able to qualitatively differentiate the fuel sensitivity of the two isomers, with n-butanol being more reactive than isobutanol across all cases. However, for each mechanism, the computational results for required intake temperature at a given combustion timing were different than that observed in experiments by 20 to 40 °C. Furthermore, the rate of change of intake temperature versus combustion timing varied across the mechanisms, which were all different than that of the experiments. Heat release rates, generation of intermediate species (OH, HO_2, H_2O_2, CH_2O, and CO), and temperature/pressure histories corresponded well with combustion timing, though significant differences existed between the mechanisms. The results indicated that kinetic mechanisms for butanol isomers do not reliably capture the ignition behavior of HCCI engines, which points to an acute development need as low temperature combustion strategies become an increasingly important part of the pathway towards engines with higher efficiencies and lower emissions.

1. Introduction

Biofuel development has gained significant interest over time due to the finite availability and carbon footprint of fossil fuels. Ethanol produced from corn or sugarcane, termed a first generation biofuel, is already in widespread use. Second generation biofuels, such as butanol, aim to alleviate competition with food sources as fuel feedstocks and reduce "well-to-wheel" CO_2 emissions [1–3]. The process to produce butanol is very similar to ethanol, and many studies have looked at butanol, in neat or blended forms, as a fuel in either spark-ignited (SI) [4–6], diesel [7–9], or Homogeneous Charge Compression Ignition (HCCI) engines [10,11]. HCCI is a low temperature combustion strategy that offers high thermal efficiencies and low NOx and soot emissions. The viability of two butanol isomers (n-butanol and isobutanol) in a HCCI engine was previously investigated, showing higher HCCI reactivity in comparison to ethanol and gasoline [12]. The research objective of this study is to assess four chemical mechanisms for butanol isomers in a single-zone engine model and compare the results against experimental findings.

Since running experiments is time consuming and costly, the availability of accurate chemical mechanisms can enable the numerical modeling of the combustion process. A well-developed mechanism should be able to capture ignition timing, heat release profiles, and temperature/pressure histories. These parameters can evaluate the mechanism's suitability to guide engine development and future experimental studies.

1.1. Homogeneous charge compression ignition

Homogeneous Charge Compression Ignition engines possess characteristics of spark-ignited engines and diesel engines, having a homogeneous mixture of fuel and air (similar to a spark-ignited engine) and compression-ignition (like a diesel engine) [13]. HCCI engines typically operate at lean equivalence ratios (less than 0.4), resulting in a low flame temperature which precipitates reduced NOx emissions in comparison to traditional internal combustion engines [14]. Chemical kinetics largely control timing and characteristics of the combustion

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Nomenclature

BDC	Bottom Dead Center	LTC	Low Temperature Combustion
CA50	Crank Angle at which 50% of the heat has been released	MON	Motor Octane Number
		NOx	Oxides of Nitrogen
CAD	Crank Angle Degree	PM	Particulate Matter
DME	Dimethyl Ether	PODE	Polyoxymethylene Dimethyl Ether
DRG	Directed Relation Graph	RON	Research Octane Number
HCCI	Homogeneous Charge Compression Ignition	SI	Spark Ignition
HR	Heat Release	TDC	Top Dead Center
HRR	Heat Release Rate	TDI	Turbocharged Direct Injection
LHV	Lower Heating Value	UHC	Unburned Hydrocarbons

event, as a temperature rise caused by the compression is used to produce the appropriate autoignition conditions [15].

One advantage of a HCCI engine is a high efficiency, the result of a high compression ratio, the elimination of throttling losses, and a shorter combustion duration [16]. Furthermore, due to low combustion temperatures, HCCI engines have less NOx emissions relative to SI and diesel engines in addition to decreased soot, or particular matter (PM) due to the lean, homogeneous air-fuel mixture.

In terms of HCCI's disadvantages, controlling the ignition event is challenging, as autoignition is largely controlled by the chemical kinetics – which is quite sensitive to temperature and composition conditions. The combustion event can be very rapid, accompanied by high rates of heat and pressure rise. Due to the resultant high ring intensity, HCCI engines tend to have a narrow operating range [17]. Cold start is another issue, as intake temperature requirements require a thermal load, which is difficult without dual-mode operation such as an engine that can operate in both SI and HCCI configurations. Lastly, increased UHC and CO emissions can come from trapped fuel/air mixture in the crevice regions of the engine during the compression stroke [18–20] and low exhaust temperatures that cannot fully oxidize the emissions. Pollutant formation can also be attributed to the thermal boundary layer along the cylinder surfaces, which causes thermal quenching [21].

1.2. Fuel Flexibility of HCCI engines

HCCI engines can also operate using a wide range of fuels including gasoline, diesel, natural gas, and some alcohols [22–24]. Several alternative fuels have also been investigated in HCCI engines, including dimethyl ether (DME) [25], ethanol [26,27], syngas [28], polyoxymethylene dimethyl ether (PODE) [29], and others [30]. Butanol in HCCI engines has been studied as a blend with primary reference fuels, ethanol, and n-heptane [31–33]. Though it has a smaller operating range in comparison to gasoline, n-butanol as a neat fuel has shown low NOx and soot emissions [34].

1.3. HCCI modeling

Modeling HCCI engines is possible using two different approaches, single-zone and multi-zone models. In the single-zone (zero-dimensional) model, it is assumed that the whole domain has uniform thermodynamic and transport properties. This method has been implemented in a number of HCCI studies for different purposes such as low temperature heat release measurement [35], ignition delay [36], NOx formation [37], ion current [38], and fuel reactivity [39]. In contrast, a multi-zone model divides the computational domain into several regions, each possessing different thermodynamic and transport properties. Therefore, the effect of crevices and near-wall thermal boundary layers can be considered. However, this method requires relatively more computational cost compared to the single-zone model. The multi-zone model has been widely employed to study detailed combustion parameters such as mean effective pressure [40], CO and UHC emissions [41], combustion duration [42], and radiation [43].

Typically, the single-zone model underpredicts CO and unburned hydrocarbon emissions, and overpredicts peak pressures due to the lack of low temperature regions near walls and crevices. However, it has been shown to predict the combustion timing if the initial temperature and species concentrations are known [44].

2. Materials & methods

2.1. Fuels

Butanol and other longer chained alcohols have significant advantages in terms of energy density, miscibility, and corrosivity in comparison with ethanol. Butanol has a four-carbon structure and depending on the position of the hydroxyl (OH) group on the carbon chain, there are four different isomers: n-butanol (1-butanol), sec-butanol, isobutanol (2-methyl-1-propanol), and tert-butanol. Sec-butanol is a straight-chain molecule with the OH group attached to an internal carbon, while n-butanol also has a straight-chain structure but the alcohol is located at the terminal carbon. Isobutanol is a branched isomer with the OH group at the terminal carbon, while tert-butanol refers to the branched isomer with the OH group at an internal carbon.

Isobutanol, n-butanol, and sec-butanol can be produced from sugars through fermentation or via the catalytic conversion of synthesis gas [45]. The remaining isomer, tert-butanol, does not exist in nature [46]. Isobutanol and n-butanol are heavily favored in terms of production from agricultural feedstocks. Of all the butanol isomers, n-butanol has been studied as an engine fuel the most. Processes that generate isobutanol from biomass resources have been studied extensively, resulting in an increased interest in its potential as a transportation fuel [47–49]. The specific properties of the fuels used in this study, alongside ethanol and gasoline, are shown in Table 1.

The butanol isomers possess an energy density closer to gasoline than ethanol. Other important advantages of butanol over ethanol include a lower heat of vaporization (improves cold start behavior), a

Table 1
Properties of the butanol isomers, ethanol, and gasoline [18,45,50]

	n-butanol	Isobutanol	Ethanol	Gasoline
Lower Heating Value (LHV) (MJ/L)	26.9	26.6	21.4	30–33
Density (kg/m³)	809.5	801.8	789.3	720–780
Research Octane Number (RON)	96	105	130	88–98
Motor Octane Number (MON)	78	94	96	80–88
Cetane number	25	–	8	0–10
Melting temperature (°C)	−89.5	−108	−114.1	–
Boiling temperature (°C)	117.7	108	78	35–200
Heat of vaporization (kJ/kg at 25 °C)	707.9	684.4	919.6	~351
Self-ignition temperature (°C)	343	415.6	434	~300
Solubility in water at 20 °C, wt%	7.7	8.7	Miscible	Negligible
Kinematic viscosity (cSt) at 20 °C	3.6	8.3	1.5	0.37–0.44