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Numerical study of formaldehyde and unburned methanol emissions of direct injection spark ignition methanol engine under cold start and steady state operating conditions

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HIGHLIGHTS

- Formaldehyde and unburned methanol emissions are numerically investigated.
- Cold start and steady state conditions are used to compare formaldehyde emission.
- Formaldehyde and unburned methanol are higher significantly for cold start mode.
- Cylinder temperature is the main factor that affects formaldehyde generation and consumption.

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ABSTRACT

The effects of overall equivalence ratio, injection timing, and ignition timing on formaldehyde and unburned methanol emissions, cylinder temperature histories, and formaldehyde emission histories of a spark ignition direct injection stratified charge methanol engine during cold start and steady state conditions were simulated using computational fluid dynamics coupling the methanol chemical kinetics reaction mechanism. The model results show that the overall equivalence ratio is less than 0.43, and unburned methanol significantly higher for cold start mode. For steady state mode, when the overall equivalence ratio is less than 0.4, formaldehyde and unburned methanol emissions increase rapidly. When the overall equivalence ratio is larger than 0.4, formaldehyde and unburned methanol emissions are very low. Formaldehyde and unburned methanol emissions for steady state mode are significantly lower than for cold start mode. For steady state mode at engine speed 1600 rpm and brake mean effective pressure 0.67 MPa formaldehyde and unburned methanol emissions are reduced 90% and 98%, respectively, compared to cold start mode at the same overall equivalence ratio (0.5). Cylinder temperature is the main factor that affects formaldehyde generation and consumption. There is a rapid decrease due to oxidation at the corresponding position of the maximum cylinder temperature, after which formaldehyde is quickly generated for any operating conditions.

1. Introduction

Energy savings coupled with reduction in emissions have been pressing global issues of the past few decades [1]. For example, the target of CO_2 emissions in the European Union is required to be below 130 g/km by 2015 for all new cars and below 95 g/km is expected to be demanded by 2021. For gasoline direct injection engine, according to EURO 6b, both the mass and the number of particulate emission must be kept below 4.5 mg/km, and 6×10^{12} #/km respectively. Euro 6c which will be enacted from 2017 demands further reduction of particulate number to 6×10^{11} #/km which is ten times stricter than that of EURO 6b [2,3]. Methanol is renewable environmentally and economically attractive alternative; and is considered to be one of the most favorable replacements for conventional fossil-based fuels [4–7]. Methanol has a high octane rating, indicating excellent antiknock performance; high latent heat of vaporization, allowing denser

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fuel-air charge; and high laminar flame speed, increasing thermal efficiency by completing combustion quicker, which also decreases heat losses from the cylinder [8,9]. However, the drawback of the use of methanol such as its low energy density, low cetane number, corrosion properties and water solubility make it difficult on the vehicle application [4,10]. Especially, the high latent heat of vaporization and longer ignition delay means methanol engines have problems with production of more unregulated formaldehyde (HCHO) and unburned methanol (CH_3OH) emissions under transient and steady state modes, and difficulties with cold starting [11–16].

Liao et al. [17] and Zhang et al. [18,19] studied the laminar burning velocities for mixtures of methanol and air at elevated temperature. They found that the laminar burning velocity was dependent on the equivalence ratio and initial temperature. The Markstein length decreased with increased initial pressure and initial temperature. Combustion pressure, mass burning rate, and burned gas temperature reached their maximum at equivalence ratio 1.1, while flame development and combustion duration were minimum at that equivalence ratio. Methanol burns fastest under rich conditions for ambient as well as high pressure [20]. The laminar flame speeds of methanol are higher than those of ethanol and the heavier alcohol flames, under fuel-rich conditions [21,22]. Wu et al. [23] studied idle lean burn characteristics of a port fuel injection spark ignition (SI) engine fueled with gasoline or methanol, and showed that the SI methanol engine had superior lean burn performance than the gasoline engine.

Chen et al. [24] reported an experimental and modeling study on the influence of methanol on premixed fuel-rich *n*-heptane flames, and found that as the equivalence ratio increased, the maximum flame temperature was reduced and the flame front shifted away from the burner surface. Formaldehyde production was promoted with the addition of methanol. Zhang et al. [25] developed a detailed oxidation mechanism for the prediction of formaldehyde emission from methanol-gasoline SI engines. Their simulation of the proposed mechanism had generally good agreement with the experimental data. At the end of the intake process, low temperature oxidation of methanol occurs in the cylinder, and some formaldehyde is generated, which increased with increasing methanol in the fuel blend. After combustion commences, formaldehyde is generated quickly, then decreases by rapid oxidation. Liu et al. [26] investigated influencing factors for formaldehyde emissions during methanol oxidation. They found the methanol incipient oxidation temperature was approximately 723 K under the test conditions. Methanol oxidation was mainly affected by temperature, with reaction time a secondary effect when the temperature exceeded 933 K. Formaldehyde first increased then decreased with temperature increase, because formaldehyde formation and oxidation are mainly affected by temperature. The critical temperature range for formaldehyde generation and oxidation was 848–882 K under the test conditions. Formaldehyde increased with increased excess air ratio. Methanol and formaldehyde emissions from the M10 engine decreased significantly when the exhaust pipe was insulated under medium and high load conditions.

Zhen et al. [27] simulated the original emissions for an SI methanol engine based on chemical kinetics including 84 reactions and 21 species. They found that produced formaldehyde was consumed quickly in the later stages of the combustion process, so very little residual formaldehyde remained after combustion. Li et al. [28] investigated the emissions of formaldehyde and unburned methanol from an SI methanol engine during cold start, and found that the methanol injection quantity per cycle, ignition timing, and methanol injection timing significantly affected formaldehyde and unburned methanol emissions. Changes in emitted formaldehyde and unburned methanol showed opposite tendencies to changes variations in methanol injected quantity per cycle, ignition timing, and methanol injection timing. Qu et al. [29] investigated regulated and unregulated emissions from a direct injection spark ignition (DISI) methanol engine under homogenous combustion and light load. They also found that changes in emitted formaldehyde and unburned methanol showed opposite tendencies to changes in methanol injection timing, ignition timing, excess air ratio, intake air temperature, and engine speed.

Advancing ignition timing, using a lean mixture, and reducing the intake air temperature can decrease formaldehyde and unburned methanol emissions, and vice versa. Agarwal et al. [30] measured unregulated emissions from a gasohol (gasoline-alcohol blend) fueled SI engine using an online high resolution Fourier transform infrared emission analyzer under steady-state engine operating conditions. They found the blend produced lower formic acid, *iso*-butane, and *iso*-pentane. Ethanol based gasohols emitted higher acetaldehyde and there were higher alcohol emissions from higher alcohol proportion gasohols.

Wang et al. [31] evaluated the effects of engine misfire on regulated and unregulated emissions from a methanol fueled engine, and its ozone forming potential. Unburned methanol emission increased by least 1.6 and 5.7 times for misfire rates of 6% and 9%, respectively. CO (carbon monoxide), toluene, xylene, and formaldehyde were the main pollutant species contributing to secondary ozone formation.

High levels of formaldehyde formation occur for SI combustion using methanol [32]. Yao et al. [33] investigated the combustion and emission formation of a dimethyl ether (DME) and methanol dual-fuel homogeneous charge compression ignition engine. The main composition of hydrocarbon emissions were unburned fuels (DME and methanol) and formaldehyde. The unburned fuels mainly arose from the piston ring crevice region, and formaldehyde from the region next to the cylinder liner.

Most previous researches have mainly concentrated on the combustion and regulated emissions of gasoline/methanol, ethanol/gasoline, and hydrogen-methanol blends; and methanol in SI engines [34–42]. However, there is increasing interest in the formation and oxidation of unregulated emissions in-cylinder. In particular, there are very limited research on the formation and oxidation of unregulated emissions in-cylinder from stratified charge direct injection spark ignition methanol engines under transient and steady state. Therefore, the unregulated emissions on methanol engine have not been adequately investigated. The aim of the current study was to simulate the effect of the overall equivalence ratio, and injection and ignition timing on formaldehyde and unburned methanol emissions in-cylinder histories, and formaldehyde emission histories of a direct injection spark ignition stratified charge methanol engine during cold start and steady state conditions. Those results should be valuable in better understanding the formation and oxidation of unregulated formaldehyde emission in-cylinder and improving formaldehyde emission for stratified charge direct injection spark ignition methanol engines under different transient and steady conditions.

2. Modeling methodology

2.1. Engine model

The modeled engine is a 1.99 L single cylinder direct injection spark ignition methanol engine which was modified from a diesel engine. The detailed specifications of engine, experimental system and fuel injection system can be found in Ref. [8]. A 7-hole, 0.45 mm, non-uniform spray line distribution nozzle was used in this study. Spark plug and injector spray line distribution are