Numerical study of hydrogen addition to DME/CH4 dual fuel RCCI engine

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Abstract
In this study, the effect of hydrogen addition to DME/CH4 dual-fuel RCCI (Reactivity Controlled Compression ignition) engine is investigated using three dimensional calculations coupled with chemical kinetics. A new reduced DME (Dimethyl Ether) oxidation mechanism is proposed in this study. With the addition of H2, the ignition time is advanced and the peak cylinder pressure is increased. The addition of hydrogen has a greater effect on the beginning stage of combustion than the later stages of combustion. The CH4 emission is reduced with the addition of H2. However, as the flame does not propagate throughout the charge, the CH4 emission is still high. The CO emission is reduced and most of the remaining CO is produced by the combustion of the premixed CH4. With the addition of hydrogen, NO emission is increased. The simulation shows that the final NOx emissions are significantly determined by the injection strategy and quantity of the pilot fuel during dual fuel operation conditions.

1. Introduction
Limited fossil fuel availability and air pollution have drawn a continuously increasing attention to the study of alternative fuels [1–4]. Natural gas is a fossil fuel, and it is not renewable. However, methane (which is the main constituent of natural gas) can be produced in renewable manners. Nature gas is a clean-burning alternative fuel for vehicles with a significant potential for reducing smoke emissions. As the renewable energy source with excellent characteristics, dimethyl ether has received considerable attention [5]. DME can be derived from many sources, including renewable materials (biomass, agricultural products and waste) and fossil fuels (coal and natural gas).

Recently, natural gas duel fuel RCCI engines have drawn a lot of attention. RCCI engine was first proposed by Rolf D. Reitz et al. [6]. The experiments showed that the US 2010 heavy-duty NOx and soot emissions regulations can be easily met without after-treatment, while achieving greater than 50% net indicated thermal efficiency. Under dual fuel conditions, the NOx emissions can be significantly reduced in comparison to normal diesel engine conditions [7,8]. Smoke or particulate emissions of the dual fuel engine are very low and even undetectable [9]. Unburned HC (hydrocarbon) emissions of dual-fuel engines are obviously higher than that of the diesel engine at part loads. At high engine loads, unburned HC emissions are comparable to those of the diesel fuel operations [10]. CO emissions tend start at a comparatively higher value at low loads, and gradually approach normal diesel engine levels with increasing equivalence ratio [11].

However, the turbulent flame speeds, flame propagation characteristics, and emissions generation characteristics of natural gas in dual fuel engine conditions are not well known. There is a demand for experimental and calculational studies...
of natural gas combustions in typical dual fuel engine operating conditions. The effects of various additions such as hydrogen (to accelerate combustion process) or EGR (Exhaust Gas Recirculation) (to reduce HC or NOx emissions) to the natural gas combustion progress are even less well known. Similarly additional experimental and numerical researches are required to investigate and predict the emission trends.

The objective of this study is to investigate the effects of hydrogen addition to DME/CH₄ dual-fuel combustion and emission characteristics. Especially the emissions generation characteristics of pure methane and methane/hydrogen blended fuels in dual fuel engine operating conditions. The potential of emission reduction of the dual fuel engine with hydrogen addition will be discussed.

2. Fuel combustion mechanism and validation

The detailed chemical reaction mechanism for the oxidation process of natural gas is GRI-Mech 3.0 [12]. GRI 3.0 consists of 53 species and 325 elementary chemical reactions with associated rate coefficient expressions. It is an optimized mechanism designed to model natural gas combustion, including NO formation and reburn chemistry. It includes the detailed oxidation reaction mechanism of hydrogen and has been widely used as the combustion mechanism of the CH₄/H₂ blended fuels. It is used as the CH₄/H₂ blended fuel oxidation mechanism in this study.

2.1. DME sub-mechanism reduction

The detailed oxidation model can represent the physical and chemical properties of the fuel better, but with the cost of increased computational complexity. Under DME/CH₄ dual fuel conditions, the main fuel is CH₄. Thus, the present study focused on the role of the CH₄, while using a reduced model for the representation of DME. The detailed oxidation mechanism of the DME [13] developed by Lawrence Livermore National Laboratory (LLNL) with 351 reactions and 79 species has been chosen as reference for reduction. This choice has been guided by the fact that the LLNL model is the most detailed mechanism and has been validated by different authors with good agreement.

The reduced mechanism is simplified in the principle that it is created by removing intermediate species with very low concentrations and associated reactions. The reaction rates of the retained reactions are the same as the master mechanism, such there is no tuning or modification of the base mechanism data.

The four steps in the present development of the reduced mechanism are as follows:

1. Identify the most essential species and reaction steps of the base mechanism using reaction rate sensitivity analysis over a wide range of operating conditions.

The sensitivity analysis was performed by multiplying the rate constant of a reaction by a factor of two (both forward and reverse rate constants). Then, in the case of the shock-tube experiments of Pfahl et al. [14], the percent change in ignition delay time compared with the baseline simulation was calculated by using SENKIN package. Senkin is a fortran computer program that computes the time evolution of a homogeneous reacting gas mixture in a closed system. The shock tube experiment was carried out using stoichiometric DME-air mixtures at two initial pressures of approximately 13 and 40 bar. A negative percent change indicates an increased overall reactivity and a positive percent change represents a decreased overall reaction rate of the system. Three different temperatures were chosen to help indicate sensitivity of each class to the onset, middle and end of the NTC region. The reactions that exhibited the highest reaction rate sensitivity at an average pressure of 40 bar are shown in Fig. 1.

At low temperatures the reactions with the most effective in promoting the overall rate of oxidation are H atom abstractions from the fuel by HO₂, OH and O₂, isomerization of

![Fig. 1 – Calculated reaction rate sensitivity.](image-url)
the CH$_3$OCH$_2$O$_2$, and the addition of CH$_3$OCH$_2$O$_2$H to O$_2$ and subsequent reactions which lead to chain branching.

At high temperature the main reactions are the decomposition reaction of H$_2$O$_2$, and the reactions of CH$_3$ and CH$_2$O with HO$_2$, and the fuel decomposition reaction, and H atom abstractions from the fuel by CH$_3$O$_2$, HO$_2$, O$_2$ and CH$_3$.

2. Eliminate unimportant species (for example, CH$_2$, CH, C$_2$H, C$_2$H$_2$, C$_3$H$_3$, C$_3$H$_6$OH, pC$_3$H$_6$OH, sc$_2$H$_4$OH, CH$_3$CO, CH$_3$CHO, CH$_3$CHO and so on) and associated reactions from the fundamental mechanism, as those species and the reactions involving them have almost no effect on ignition delay time.

3. Run SENKIN to simulate the shock-tube experiments of Pfahl et al. to obtain the species concentrations and the experimental data from flow reactor experiments are modeled to check the species profiles using the closed homogeneous reactor model under adiabatic conditions. All the experiments were carried out by Pfahl et al. [14].

Generally, the ignition time definitions are various, such as peak OH, peak CO or temperature inflection. As the resulted ignition times are very similar, all the calculated ignition times are based on temperature inflections in this study. Fig. 2 presents the comparisons of the reduced DME oxidation model predictions of ignition delays (lines) with experimental data (dots) [14]. The selected conditions are stoichiometric DME-air mixtures with two initial pressures of approximately 13 and 40 bar. Good agreement between the model predictions and the experimental data are shown over the wide range of conditions considered.

The reduced DME mechanism described above is compared with the pyrolysis and oxidation experiments in the variable pressure flow reactor (VPFR). On-line, continuous, extractive sampling in conjunction with Fourier Transform Infra-Red, electrochemical analyzer (for O$_2$), non-dispersive infrared (NDIR) analyzers (for CO and CO$_2$), and selective detector (for H$_2$) were performed to quantify species at specific locations along the axis of the turbulent flow reactors [13]. Relative times of various axial positions to one another were defined by integrating the velocity distribution function along the axial coordinate. However, in regions of high-concentration gradients, axial diffusion is not negligible. Furthermore, the finite-rate mixing of fuel and oxidizer and, potentially, small recirculation zones near the mixing region may have a significant effect.

Finally, a reduced mechanism of DME with 27 species and 46 reactions is obtained in this study, as shown in Table 1.

### 2.2. DME sub-mechanism validation

Normally, before using reaction mechanisms in complex simulations, the validation of the reaction mechanism should be taken into account over a wide range of conditions. The results from such tests with our reduced mechanism are presented in this study. The experimental data considered are all from well defined laboratory experiments, which include ignition times from shock tubes and species profiles from flow reactors.

The SENKIN code is used for the mechanism validation modeling. The shock tube experiments are modeled to check the ignition delays, assuming constant-volume, homogeneous, adiabatic conditions behind the reflected shock wave; the flow reactor experiments are modeled to check the species profiles using the closed homogeneous reactor model under adiabatic conditions. All the experiments were carried out by Pfahl et al. [14].

<table>
<thead>
<tr>
<th>Number</th>
<th>Reaction</th>
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</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>CH$_4$+OH $\rightarrow$ CH$_3$+H$_2$O</td>
<td>24</td>
</tr>
<tr>
<td>2</td>
<td>CH$_3$+CH$_2$O $\rightarrow$ HCO + CH$_4$</td>
<td>25</td>
</tr>
<tr>
<td>3</td>
<td>CH$_3$+HO$_2$ $\rightarrow$ CH$_3$O + OH</td>
<td>26</td>
</tr>
<tr>
<td>4</td>
<td>CH$_3$+HO$_2$ $\rightarrow$ CH$_3$O + O$_2$</td>
<td>27</td>
</tr>
<tr>
<td>5</td>
<td>CH$_3$+O$_2$ $\rightarrow$ CH$_3$O + OH</td>
<td>28</td>
</tr>
<tr>
<td>6</td>
<td>CH$_3$O + CH$_3$ $\rightarrow$ CH$_3$O + CH$_4$</td>
<td>29</td>
</tr>
<tr>
<td>7</td>
<td>CH$_3$O+CH$_3$(M) $\rightarrow$ CH$_3$ + (M)</td>
<td>30</td>
</tr>
<tr>
<td>8</td>
<td>HCO + O$_2$ $\rightarrow$ CO + HO$_2$</td>
<td>31</td>
</tr>
<tr>
<td>9</td>
<td>HCO + M $\rightarrow$ H + CO + M</td>
<td>32</td>
</tr>
<tr>
<td>10</td>
<td>CH$_3$O + HO$_2$ $\rightarrow$ HCO + HO$_2$</td>
<td>33</td>
</tr>
<tr>
<td>11</td>
<td>CH$_3$O + OH $\rightarrow$ HCO + H$_2$O</td>
<td>34</td>
</tr>
<tr>
<td>12</td>
<td>CH$_3$O + H $\rightarrow$ HCO + H$_2$</td>
<td>35</td>
</tr>
<tr>
<td>13</td>
<td>CH$_3$O(+M) $\rightarrow$ CH$_3$O + H(+M)</td>
<td>36</td>
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<tr>
<td>14</td>
<td>HO$_2$+OH $\rightarrow$ H$_2$O + O$_2$</td>
<td>37</td>
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<td>15</td>
<td>CH$_3$O$_2$+M $\rightarrow$ CH$_3$+O$_2$+M</td>
<td>38</td>
</tr>
<tr>
<td>16</td>
<td>CH$_3$O$_2$H $\rightarrow$ CH$_3$O + OH</td>
<td>39</td>
</tr>
<tr>
<td>17</td>
<td>CO + OH $\rightarrow$ CO$_2$+H</td>
<td>40</td>
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<tr>
<td>18</td>
<td>CO + HO$_2$ $\rightarrow$ CO$_2$+OH</td>
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<tr>
<td>19</td>
<td>OH + OH(+M) $\rightarrow$ H$_2$O$_2$(+M)</td>
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<tr>
<td>20</td>
<td>H + O$_2$ $\rightarrow$ O + OH</td>
<td>43</td>
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<tr>
<td>21</td>
<td>H + O$_2$(+M) $\rightarrow$ HO$_2$(+M)</td>
<td>44</td>
</tr>
<tr>
<td>22</td>
<td>H$_2$O$_2$+O$_2$ $\rightarrow$ HO$_2$+HO$_2$(dupl)</td>
<td>45</td>
</tr>
<tr>
<td>23</td>
<td>H$_2$O$_2$+O$_2$ $\rightarrow$ HO$_2$+HO$_2$(dupl)</td>
<td>46</td>
</tr>
</tbody>
</table>
all result in uncertainty in the definition of the absolute “zero time” for flow reactor experiments. In this study, the points of 50% observed fuel disappearance were matched to compare the calculation and experiment. Fig. 3 gives the species profiles from VPFR experiments (dots) [13] against model predictions (lines). The calculation was started under stoichiometric condition with atmospheric pressure at a temperature of 1084K. The model predicts well the flow reactor data for major species (DME, H2O, CO) and even for trace intermediates (methane, formaldehyde, ethylene and ethane).

3. CFD package and sub-models

The KIVA-3V simulation package was used for the RCCI engine simulations. It is a transient, three dimensional, multiphase, multi-component code for the analysis of chemically reacting flows with sprays, which has been under development at the Los Alamos National Laboratory. Since detailed oxidation mechanism will be used to simulate fuel combustion, the CHEMKIN chemistry solver was integrated into the KIVA-3V code for solving the oxidation process during multi-dimensional engine simulations. Then, the chemistry and flow solutions were coupled. It should be noted that the convection and diffusion transport between different computational cells were modeled by the conventional k-ε turbulence model.

The TAB (Taylor Analogy Breakup) model is used to simulate the spray droplet aerodynamic breakup. This model is based upon a taylor analogy between an oscillating and distorting droplet and a spring-mass system. This model is used in conjunction with the reitz atomization method. It predicts the spray angle in addition to the state of oscillation and distortion.

The Partially Stirred Reactor (PaSR) combustion model was used to model the chemistry/turbulence interaction process. In the PaSR approach, a computational cell is distinguished into two different zones, reaction zone and no reaction zone. In the reaction zone all compositions are assumed to be perfectly mixed with each other, called perfectly stirred reactor. Three average concentrations (in mean molar density) are presented in the reactor: the mean molar density of the feed \( c_0 \); the concentration in the reaction zone \( c \); the concentration at the exit of the reactor \( c_1 \).

The whole combustion process is according to the following two steps. First, the concentration in the reaction zone changes from \( c_0 \) to \( c \) due to the chemical reaction. Second, the reaction mixture \( c \) is mixed with the no reaction mixture \( c_0 \) by turbulence, which results in the averaged concentration \( c_1 \).

The combustion in the reactor is a sequential process where mixing is followed by chemical reaction and that for a stationary regime the rates of the individual steps and of the overall process must be equal. It seems quite clear that the reaction rate should be proportional to the ratio of the chemical reaction time \( \tau_{\text{chem}} \) to the total combustion time in the reactor. The total time of the combustion process is a sum of the mixing \( \tau_{\text{mix}} \) and reaction times \( \tau_{\text{chem}} \).

\[
k = \frac{\tau_{\text{chem}}}{\tau_{\text{chem}} + \tau_{\text{mix}}}
\]

The mixing time \( \tau_{\text{mix}} \) characterizes the exchange process between reaction mixture and no reaction mixture. In this paper the mixing time was obtained from the k-ε equation, \( \tau_{\text{mix}} = c_{\text{mix}} k/\varepsilon \), the model constant \( c_{\text{mix}} \) was set to 0.015. This
constant can be varied between 0.001 and 0.3, depending on the flow conditions. The reaction time was derived from the laminar reaction rate (Arrhenius reaction rate). Thus, the overall reaction rate \( \omega \) and the homogeneous reaction rate \( \sigma \) of this computational cell have the following relationship:

\[
\frac{C_1 - C_0}{dt} = \omega = k\sigma
\]

If \( t_{\text{mix}} \) equals zero, the combustion model is reduced to the quasi-laminar approach. If \( t_{\text{mix}} \) dominates, the reaction rate is reduced to the Magnussen-Hjertager eddy breakup rate. Then, the overall reaction rate in the PaSR model could be also regarded as the average of the quasi-homogeneous rate and the turbulent mixing control rate.

A four-stroke single cylinder Gardner compression ignition engine was modeled in this work. The engine specifications are shown in Table 2. Further details can be found in [15].

In the dual-fuel tests, natural gas/hydrogen blended fuels were supplied directly into the inlet manifold, where they were inducted through one flow meter under the engine’s own suction along with air. The amount of pilot fuel was fixed at a flow rate meant for 0.1 MPa brake mean effective pressure (BMEP) at 1000 r/min during normal engine operation. The pilot DME was supplied to the engine’s high-pressure pump in the liquid state from a commercial high-pressure tank, pressurized with a nitrogen gas source at 5.2 bar. The pilot fuel was directly injected into the cylinder at the same injection timing (24.5° before top dead centre, BTDC).

For dual-fuelling operation, the load was controlled by adjusting the flow rate of natural gas/hydrogen blended fuel inducted by the engine. Three natural gas/hydrogen blended fuels (containing 0%, 20%, and 40% of hydrogen in volume) were used in this study. The experiment process was as follows: first, the pure natural gas flow rates started at 0 l/min (0.1 MPa BMEP), then the load would be increased to 0.378 MPa BMEP by increasing the natural gas flow rate appropriately [15]. This is the base condition. After that, the pure natural gas was changed to two natural gas/hydrogen blended fuels. The flow rates of the blended fuels started at 0 l/min (0.1 MPa BMEP), then the flow rates were increased until the natural gas flow rates equal to the base condition. In this case, the quantity of the pilot fuel and natural gas are all the same. But the quantity of the hydrogen was changed.

The three dimensional computational grid used in this study is shown in Fig. 4. In order to reduce computational burden, a 90° sector mesh with periodic boundaries was used for the spray and combustion simulations. In the vertical and axial directions, the mesh is fairly coarse. To determine the sensitivity of results to the mesh resolution, we compared results with those using a finer mesh. The results using the two meshes were quite similar, with CA50 within 1.0°–2.0° for the cases modeled. Therefore, the coarser mesh has been used for all the simulations. The computations were run from IVC (intake valve close) to EVO (exhaust valve open). Chemistry oxidation calculations were called from −50° ATDC (after top dead center) to 75° ATDC. Beyond these crank angles, it was found that the chemistry was negligible for the cases simulated. During the simulation, the quantity of the pilot DME and the premixed natural gas was kept constant. The quantity of the hydrogen was increased according to the mole fraction of the natural gas.

### Table 2 – Engine specifications.

<table>
<thead>
<tr>
<th>Specification</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bore</td>
<td>107.95 mm</td>
</tr>
<tr>
<td>Stroke</td>
<td>152.4 mm</td>
</tr>
<tr>
<td>Displacement volume</td>
<td>1394.8 CC</td>
</tr>
<tr>
<td>Compression ratio</td>
<td>13:1</td>
</tr>
<tr>
<td>Inlet valve opening (IVO)</td>
<td>10° BTDC</td>
</tr>
<tr>
<td>Inlet valve closing (IVC)</td>
<td>40° ABDC</td>
</tr>
<tr>
<td>Exhaust valve opening (EVO)</td>
<td>50° BBDC</td>
</tr>
<tr>
<td>Exhaust valve closing (EVC)</td>
<td>15° ATDC</td>
</tr>
<tr>
<td>Injection timing</td>
<td>24.5° BTDC</td>
</tr>
<tr>
<td>No. of injector nozzle holes</td>
<td>4</td>
</tr>
<tr>
<td>Injector throat diameter</td>
<td>220 µm</td>
</tr>
<tr>
<td>Injector opening pressure</td>
<td>16.2 MPa</td>
</tr>
<tr>
<td>Engine speed</td>
<td>1000 r/min</td>
</tr>
</tbody>
</table>

Fig. 5 illustrates the calculated cylinder pressures with different H₂ additions. The experiment data from Namasiyavam’s experiment are also plotted in this figure. It shows good agreement between the computed and measured cylinder pressures. With the addition of H₂, the ignition time is advanced and the peak cylinder pressure is increased. The addition of hydrogen promotes the production of hydroxyl (OH) radicals, which increases the combustion reactivity during the flame development period [16]. Moreover, the higher flame speed of hydrogen speeds up combustion [17–21], resulting in more fuel being burnt in the time available.

Fig. 6 presents the CH₄ mole fractions with different H₂ additions. The mole fractions of CH₄ were increased at the beginning of the combustion process. That is because the oxidation of DME can produce some CH₄. Normally, the unburned HC emissions of the natural gas fueled dual-fuel engines are obviously higher than that of the diesel engines [22] at low to intermediate loads. This is caused by the unburned natural gas (mostly methane) surviving to the exhaust. There are three major factors contributing to this
phenomenon. Firstly, the gaseous fuel and air mixture being too fuel-lean for combustion, the conditions of temperature and pressure and their change make propagation of the expanding flame ball increasingly difficult. Secondly, part of the premixed natural gas is forced into the crevices of the piston ring and dead zone of the combustion chamber during the compression strokes, which escapes from combustion. Finally, as the combustion of the natural gas-air mixture usually begins in the expansion stroke, flame quenching may be a contributing factor [23]. With the addition of hydrogen, the consumption rate of CH₄ is increased, especially in the initial 10 to 15 crank angles of the combustion period. However, at the latter combustion stages, the addition of hydrogen has no obvious effect on the combustion process.

Fig. 7 gives the variation of CO mole fractions with different H₂ mole fractions. Generally, the CO emissions of the natural gas dual-fuel engines are significantly higher than that of the normal diesel engines, especially at part load conditions. As shown in Fig. 7, CO species are produced at the initial combustion stage, and then part of them is consumed by oxidation reactions. With the addition of hydrogen, CO emission is reduced. That is because the addition of H₂ advances the ignition time and increase the combustion temperature, which promotes the oxidation of CO.

The profiles of NO mole fractions with different H₂ mole fractions are shown in Fig. 8. Usually, in dual-fuel engines, the flame temperatures are less than that in diesel engines, since most of the fuel burns under lean premixed conditions. So the NOx emissions of the dual fuel engines tend to be lower than that of the diesel engines. Moreover, the combustion of the natural gas-air mixture usually begins in the expansion stroke, which suppresses the temperature increase and freezes NOx production chemistry. In addition, other work [24] reports that the combustion of the pilot fuel produced most of the total NOx emissions of the dual-fuel engine. Therefore, the injection strategy and quantity of the pilot fuel significantly determine the final exhaust NOx emissions during dual fuel operation conditions. With the addition of hydrogen, NO emission is increased. That is because the addition of H₂ advances the ignition time and increases the combustion temperature, which promotes the production of NO.

To further understand the combustion process, in cylinder images from the modeling work are presented in Figs. 9–12, where two cut planes coincident with the spray axis colored

Fig. 9 – Computational temperature.

Fig. 10 – Computational CH4 mole fraction.

Fig. 11 – Computational CO mole fraction.

Fig. 12 – Computational NO mole fraction.

by temperature, mass fraction of CH₄, mass fraction of CO and mass fraction of NO are shown.

Figs. 9 and 10 indicate that the premixed pilot DME ignites first, then after a few crank angles, the gaseous fuel (CH₄) entrained within its envelope starts to burn and then the flame propagates through the CH₄/air mixture. However, the flame does not propagate throughout the charge. With the addition of hydrogen, the ignition time is advanced and the combustion region is enlarged. CO is formed even before the ignition, and then some of them are consumed by oxidation reactions, especially in the DME combustion region (high temperature region). As shown in Fig. 11, most of the remaining CO is produced by the combustion of premixed CH₄. NOₓ is formed later after the combustion when the combustion temperature is sufficiently high, as revealed in Fig. 12. And the NOₓ formation region is mainly within the pilot fuel evaporation area, which confirms that the combustion of the pilot fuel produces most of the total NOₓ emissions of the dual-fuel engine.

5. Conclusions

The effect of hydrogen addition to DME/CH₄ dual fuel engine is studied using three dimensional calculations coupled with the detailed chemistry mechanism. The mole fraction profiles of the primary species and the in cylinder images from the modeling work are presented. The main results are summarized as follows:

1. A new reduced DME oxidation model is proposed in this study. It is capable to simulate the ignition and combustion process of DME.
2. With the addition of H₂, the ignition time is advanced and the peak cylinder pressure is increased.
3. The addition of hydrogen has a greater effect on the beginning stages of combustion than in later stages of combustion.
4. With the addition of hydrogen, the CH₄ emission is reduced. As the flame does not propagate throughout the charge, the CH₄ emission is still high.
5. With the addition of hydrogen, CO emission is reduced. Most of the remaining CO is produced by the combustion of premixed CH₄.
6. With the addition of hydrogen, NO emission is increased. The injection strategy and quantity of the pilot fuel significantly determine the final exhaust NOₓ emissions during dual-fuel operation conditions.

REFERENCES


