

Characteristics of Formaldehyde (CH_2O) Formation in Dimethyl Ether (DME) Spray Combustion Using PLIF Imaging

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ABSTRACT

Recognition of Dimethyl Ether (DME) as an alternative fuel has been growing recently due to its fast evaporation and ignition in application of compression-ignition engine. Most importantly, combustion of DME produces almost no particulate matter (PM). The current study provides a further understanding of the combustion process in DME reacting spray via experiment done in a constant volume combustion chamber. Formaldehyde (CH_2O), an important intermediate species in hydrocarbon combustion, has received much attention in research due to its unique contribution in chemical pathway that leads to the combustion and emission of fuels. Studies in other literature considered CH_2O as a marker for UHC species since it is formed prior to diffusion flame. In this study, the formation of CH_2O was highlighted both temporally and spatially through planar laser induced fluorescence (PLIF) imaging at wavelength of 355-nm of an Nd:YAG laser at various time after start of injection (ASOI). The injection pressure was kept at 750 bar using a single-hole injector with diameter of 180 μm . The formation of CH_2O is also correlated with time of ignition and flame region using other optical diagnostics including photodiode and natural flame luminosity, respectively. Additionally, kinetic modeling and CFD were used in order to explain further and confirm the combustion process and species formation (particularly, CH_2O) in the DME flame. Both experimental and numerical results showed that CH_2O is formed mostly prior to flame ignition at upstream region before liftoff length.

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INTRODUCTION

Reducing emissions from engine exhaust gas including particulate matter (PM) and NO_x still remains as the main target to achieve environmental friendly combustion in the automobile. Compression ignition (CI) engines is known to be beneficial since it does not require external ignition source from spark or glow plug but rather uses auto-ignition behavior of high cetane fuel such as diesel to provide energy by combustion for engine operation. However, CI engine also potentially produces significant amount of soot due to heterogeneous mixture of fuel and air which often results in locally fuel rich area inside the combustion chamber. While there are new combustion technologies to reduce emissions in order to meet strict environmental regulation for emission such as after-treatment device for emission or NO_x , alternative fuels such as natural gas, liquefied petroleum gas (LPG) fuel are constantly being considered as the solution for our future energy generations.

As one of the alternative fuels, Dimethyl Ether (DME) has received great attraction from research and industrial community recently. Almost no soot (or smokeless flame) is achieved through the combustion of DME due to no C-C bond in its chemical formula and high oxygen content (34.8% in mass) as shown in Table 1 along with

other properties in comparison with diesel fuel. Shorter ignition delay is also expected due to high cetane number of DME (>55) compared to that of diesel (40-55). High cetane number is an important factor in fuel ignitability, especially in compression-ignition engine. Having higher cetane number than traditional diesel fuel, DME provides short ignition timing during compression stroke, therefore enables engine operation with ease. DME also has low boiling point which makes it vaporize almost immediately after injection [1]. The remaining challenges in using DME as fuel in the fuel injection system are its low lubricity and low viscosity which could potentially lead to wear problems and higher required compression work. The lubricity issue, however, can often be improved by the addition of Lubrizol, Infineum R655 or other lubricity enhancement [2].

Despite the remaining challenges in handling DME as fuel in FI system due to its physical characteristics, DME stands out as a very promising fuel in CI application when being liquefied at pressure of 8 bar or higher for liquid fuel injection, similar to diesel. DME can be injected at larger amount of fuel compared to diesel due to its lower heating value by increasing the duration, injection pressure, or both together. In general, DME is an environmental friendly with production of ultra-low soot emission

from its combustion, and can be produced from common synthesis gas (e.g. methanol) or non-petroleum feedstocks (e.g. coal, biomass, or natural gas).

Table 1. Fuel properties comparison for DME and diesel

Property	DME	Diesel
Chemical formula	CH ₃ OCH ₃	C8 to C25
C/H ratio	0.337	0.516
Oxygen content (% mass)	34.8	0
Critical temperature (K)	400	708
Critical pressure (MPa)	5.37	3
Vapor pressure at 20 °C (kPa)	530	<<1
Boiling temperature (°C)	-25	150-380
Liquid density (kg/m ³)	660	800-840
Liquid viscosity (kg/ms)	<0.1	3
Lower heating value (MJ/kg)	28.43	42.5
Cetane number	55-60	40-55
Auto-ignition temperature (K)	508	523
Stoichiometric A/F ratio	9	14.6
Enthalpy of vaporization (kJ/kg)	460	250

There have been numerous research on DME as an alternative fuel to establish extensive data for fuel properties [3]; to study the kinetic mechanism and chemical pathway [4, 5, 6]; and, to investigate its spray and combustion feature in direct injection (DI) system [7, 8]. Also, the spray and combustion of DME have been studied in combustion vessel to provide visualization and measurement of its spray structure, flame luminosity such as those studies of [9, 10]. There has been small number of studies on the emission of DME flame since it is known to have low chance to produce large hydrocarbon such as soot particle, polycyclic aromatic hydrocarbon (PAH), or soot precursors (e.g. acetylene, benzene) [11, 12]. However, some intermediate species produced during combustion of hydrocarbons such as formaldehyde (CH₂O), are considered to be harmful to the environment and human health. Formaldehyde has also been known in diesel spray combustion as the intermediate species occurring during the first-stage of ignition when there are initial weak chemiluminescence and fluorescence of some species such as OH*, CH₂O, CH* characterizing the premixed combustion event [13]. CH₂O in diesel combustion was also visualized using planar laser-induced fluorescence (PLIF) in combustion vessel simultaneously with OH chemiluminescence imaging in previous study of Pickett et al. [14]. They showed that CH₂O is formed upstream, prior to lift-off length, but is consumed downstream by high temperature regions of diffusion flame. In a DME reaction mechanism, CH₂O plays an important role in the chemical pathway of fuel oxidation process: it is formed from methoxymethyl (CH₂OCH₃) decomposition [4] and is consumed later to form carbon

dioxide through basic combustion path (CH₂O→HCO→CO→CO₂) [15]. Therefore, it is important to investigate the formation of formaldehyde and its role in the chemical reaction pathway of DME combustion as well as to correlate its profile with commonly known combustion characteristics (e.g., ignition, flame location).

In order to investigate the formation of formaldehyde, visualization such as those assisted by the experimentation in a combustion vessel becomes very useful in both temporal and spatial manners [14, 16, 17]. Therefore, the current study was performed firstly using a hydraulically activated electronic unit injector (HEUI) fuel injection system coupled into combustion research facility at Michigan Tech. A more detailed description of the combustion vessel can be obtained from ref. [16, 18]. Images of the combustion luminosity and species formation such as formaldehyde captured from natural flame luminosity and laser diagnostic technique, respectively, were then processed and analyzed. Important combustion characteristics such as ignition delay, developing structure of flame, and cool-flame combustion (characterized by CH₂O formation region) were revealed in DME flame. Ambient temperature was also varied in this study, in order to observe the change in DME ignition from low to high temperature range. This is because, like most of the fuels, the autoignition characteristic of DME relies heavily on the given ambient temperature since there exists competition of different reaction chain-branching or chain-propagating from low to intermediate temperature which was described analytically by recent study of Beckmann et al. [19]. They aimed to provide the predictive tool for ignition delay in two stages of DME combustion including intermediate temperature regime. Some previous studies have attempted to improve the overall reaction model of DME for high temperature condition [20] combining with existing DME chemistry [21, 22, 23]. Finally, as part of this work, a CFD simulation was developed and validated with experimental data as a predictive tool that is capable of providing more insights on the combustion of DME such as profile of multiple species formation during spray and combustion event. Here, the predicted formation of species such as CH₂O from the original kinetic mechanism provided valuable information onto understanding chemical pathway inside DME flame. The overall objective of this study is to achieve an understanding of formaldehyde formation and its characteristics in DME reacting spray by combining both experimental data and image with CFD results. It was found that CH₂O formation is linked strongly with flame ignition timing: CH₂O occurs before ignition in both experimental and numerical approaches. There was absence of CH₂O in high temperature regions of DME flame which implies that the low-temperature reaction prior to diffusion flame is most favorable for the formation of CH₂O.

EXPERIMENTAL SETUP AND TEST CONDITION

1. Fuel Injection System

The current experiment employed an intensified injector that uses high oil pressure to intensify the fuel pressure with ratio of 1:9.9. The overall fuel injection system is shown in Figure 1. The injector

is based on the current 2014MY Navistar injector utilized in the medium duty diesel market sector with injection pressure capability up to 2900 bar. Much of the DME studies have been reported at injection pressures ranging from 300-500 bar, with literature studies showing improved fuel economy with increased injection pressure due to better mixing and atomization [2]. This type of injector is referred to as hydraulically activated electronic unit injector (HEUI). The advantage of HEUI injector lies on the minimum exposure of the fuel (DME) to the injector nozzle alone due lower viscosity and lubricity of DME can potentially damage the physically moving parts. The injector is driven by two iPOD coil power EFS drivers to control the opening and closing coil events that are responsible for the start and end of the injection event. In this study, the injection duration (energizing time) was kept to 2.0 ms between the start of open coil and the start of close coil. The oil pressure is built up on a common rail to amplify the piston hydraulically and provide high pressure at the injector fuel outlet. Oil is then returned to 10-gallon oil reservoirs which provide very sufficient oil supply for most of desirable injection configuration (injection pressure, duration). Liquid DME fuel was achieved by keeping the pressure of the fuel supply line to the injector at 0.8 MPa using nitrogen gas. The injector nozzle was manufactured to achieve single-hole nozzle with diameter of 180 μm . Actual injector nozzle's diameter was also measured through scanning electron microscope (SEM) imaging with value of 174.2 μm .

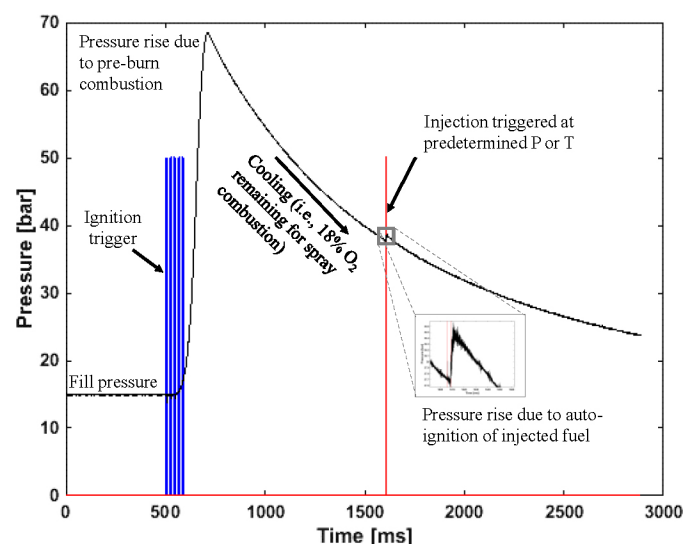


Figure 2. Example of pressure profile inside combustion chamber: pre-burn combustion, cooling phase, injection event at high temperature (pre-defined by pressure base).

The injection pressure was kept constant throughout the experiment at 750 bar with 2.0 ms energized injection duration. The injection pressure was monitored and controlled with only ± 0.3 bar fluctuation from set pressure. Note that the injection pressure was estimated based on the monitoring oil pressure and the intensification (10 times). The test condition is summarized in Table 2 with different ambient temperature by simply changing the time of injection during the cooling phase of products from pre-burn combustion.

Table 2. Experimental condition for DME reacting spray

Parameter	Value
Fuel	DME
Nozzle Diameter [μm]	180
Injection Pressure [bar]	750
Ambient Composition	18% O_2
Ambient Temperature [K]	750, 900, 1100
Ambient Density [kg/m^3]	14.8
Fuel Temperature [K]	383
Energizing Duration [ms]	2.0

Formaldehyde (CH_2O) radical was excited at the wavelength of 355 nm Nd:YAG laser (Continuum Surelite III) with a 10 ns laser pulse-width. Other similar studies have been performed to visualize CH_2O in spray combustion using the same excitation wavelength [14, 17, 25]. The species fluorescence was captured through the intensified CCD camera (DiCam Pro) shown in Figure 3 at several ASOI timing to visualize the formation and consumption of CH_2O over time. The camera was equipped with 105 mm UV lens f/4 (250-650 nm transmission) and band-pass filter 400 nm centered, 20 nm FWHM. Camera gate was set at 200 ns covering the time when laser is activated. The time setting of camera and laser triggering was verified using a high sampling rate oscilloscope, so that the ICCD gate time was able to cover fully the formaldehyde excitation period. The resolution of the image is 1280 x 1024 pixels with the resolution scale of 0.091 mm/pixel. Several studies have shown the use of optical filter to capture formaldehyde fluorescence such as 385-450

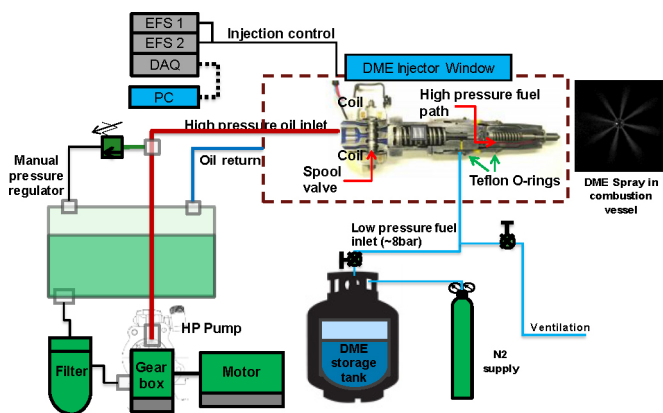


Figure 1. Diagram of fuel injection system integrated with DME injector window with the sample spray image of multi-hole injector nozzle in MTU combustion vessel

2. Constant Volume Combustion Vessel & Optical Diagnostics

The single-hole HEUI injector system was integrated into the current combustion vessel research facility. Baseline condition of the experiment consists of 18% O_2 ambient composition, density, ρ , of 14.8 kg/m^3 with ambient temperature, T_{amb} , of 900 K. This ambient condition was achieved by igniting the pre-burn mixture of C_2H_2 , H_2 , O_2 , CO_2 using electrode with turbulence controlled by a rotating fan inside the vessel for homogeneous mixture. The fuel injection event inside combustion chamber was demonstrated in sample pressure profile shown in Figure 2 at the time after combustion from pre-burn mixture was cooling down. Detailed description of the combustion vessel (CV) can also be found in Ref. [24].

nm [14, 17], UG5 Schott filter and 500 nm short-pass filter [26]. Collin et al. [27] showed the peak intensity of CH_2O at about 420–430 nm. Therefore, it is believed that the chosen filter was appropriate to obtain sufficiently formaldehyde signal while avoiding elastically scattered light or reflection from the laser excitation.

Flame luminosity from the combustion of DME was captured through a high-speed camera (Photron Fastcam SA 1.1) at 40,000 fps with the exposure time of 24.33 μs . The long exposure time was necessary to detect weak signal from DME flame, but still avoiding any image saturation from the combustion signal. The resolution of the images was 512 pixel (width) by 256 pixel (height). This allows to see a full development of the spray from injector nozzle to the opposite CV window where spray or combustion impinges on eventually. An 85-mm Nikkor lens with $f/1.8$ was integrated with high-speed camera to provide the final projected view with the scale of 0.187 mm/pixel. The high-speed camera was set up in a similar manner as the intensified CCD camera as shown in Figure 3.

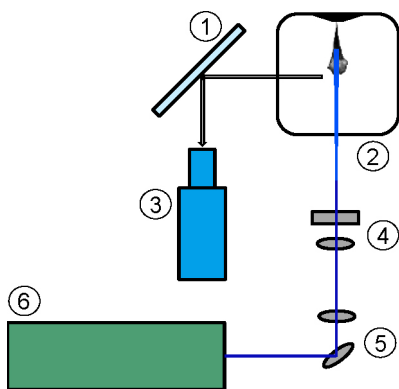


Figure 3. Optical arrangement for PLIF diagnostics of formaldehyde for combustion vessel experiment: 1. Reflecting mirror; 2. Combustion vessel with side window coupled with injector and fuel injection system; 3. ICCD camera with bandpass filter (400 nm centered, 20 nm FWHM) for CH_2O visualization (or HS camera with no filter required for natural luminosity image capturing); 4. Focused and cylindrical lens; 5. Dichroic mirrors; 6. Nd-YAG laser

RESULTS AND DISCUSSION

1. DME Flame Development and Ignition

After being injected into a hot and oxidizer available environment, DME combusts with a relatively low intensity flame. Even though the HS camera was set with maximum exposure time (24.3 μs) according to the current 40,000 frame per second rate, the start of DME combustion was not able to be identified easily by the visualization of the image due to low to moderate light at earlier stage of the combustion. An example of natural luminosity image of DME combustion is shown in Figure 4 with initial flame developed into a hot “ball”. Although through certain digital image processing, the time and location of the ignition can be achieved through intensity integration method [16], it was decided to use a photodiode (Thorlab DET36A) to detect the time of ignition in this experiment. The photodiode (PD) has fast rise time of 1.0 ns with 50 mm Nikkor lens $f/1.8$ placed in front in order to have a full view of the combustion vessel window where any luminosity from the combustion of DME could be captured. From Figure 4, the liquid signal can also be seen. This was because a LED

light was located to illuminate the near nozzle region to capture liquid droplet scattering signal, therefore acquiring the start of the injection. More importantly, the photodiode is not sensitive to liquid signal as confirmed from the recorded signal over time. Therefore, it is believed that only combustion signal was obtained via PD. The ignition delay is then calculated as the time difference between start of injection (by liquid signal from HS images) to PD signal rise. The calculated ignition delay therefore has an error of only $\pm 25 \mu\text{s}$ due to the current camera frame rate. It is also worth to mention that ignition delay can be calculated from pressure curve that is measured by pressure transducer. There exists a predetermined threshold for the rise of heat release rate (above 0.03 bar) that is processed from pressure inside the constant volume chamber for diesel combustion to determine the time of ignition [28]. However, due to uncertainty in processing the pressure profile (e.g. signal noise, post-process data filtering), this approach was not applied in this study. Therefore, the photodiode remains as the main determination for the time of ignition throughout the experiment.

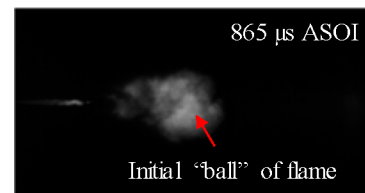


Figure 4. Sample high speed luminosity image of DME flame at 865 μs ASOI with ambient condition of 900 K, 18% O_2 , 14.8 kg/m^3 .

The photodiode signal is shown in Figure 5 for ambient temperature of 900 K. The opening and closing coil signals were overlapped on the plot. The open coil current profile is started at 0 ms, while actual injection is initiated at 1.5 ms after triggering of FI system at 750 bar injection pressure. This was determined from HS images of DME's liquid signal. The ignition delay from photodiode is approximately 0.7 ms (averaged from 7 runs).

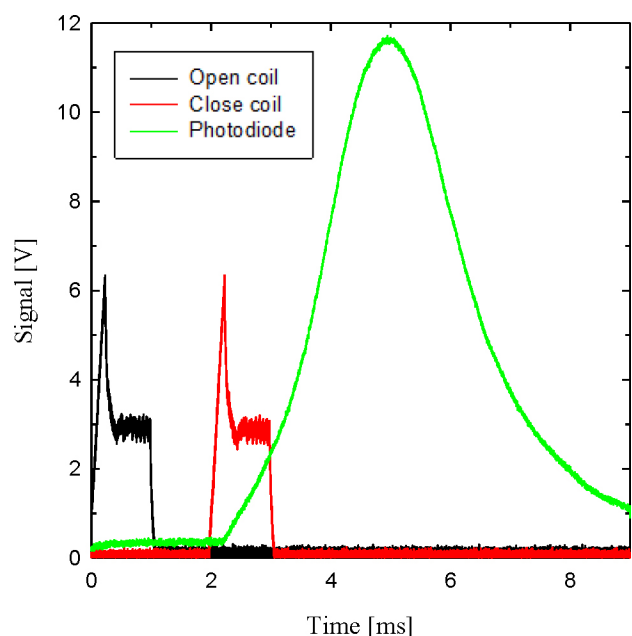


Figure 5. Photodiode signal and open/close coils time profile. Test condition was injection pressure of 750 bar, injection duration of 2.0 ms, ambient density of 14.8 kg/m^3 , ambient temperature of 900 K.