Optimization of Synthetic Oxygenated Fuels for Diesel Engines

Investigators

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Abstract

During the past year, a systematic and fundamental experimental study of combustion and soot production of various oxygenated molecules was carried out using oxygenates from the main functional families: ethers, esters, aldehydes, alcohols and ketones. The experimental studies provide data on ignition delay, soot yields and soot induction times, and detailed species evolution profiles for the fuel and major intermediate and product species. The results from the experimental studies have been compared with predictions from detailed reaction mechanisms and areas where mechanism improvements are required were identified.

To assess the effectiveness of oxygenated fuels in reducing pollutant emissions in Diesel engines and to optimize the Diesel engine process for the particular fuel characteristics, a limited set of engine experiments was carried out and methods for threedimensional numerical simulations of Diesel engine combustion were developed. The simulations permit a more extensive investigation of effects of engine design and operating conditions on performance and emissions. For the simulations, we are developing a state-of-the-art computational tool which is capable of performing Large Eddy Simulation (LES) of turbulent combustion in Diesel engines. The code has been validated by comparison of code predictions with available experimental data on unsteady flows.

Introduction

The objectives of this project are to develop novel oxygenated fuels and to establish the chemical kinetic models required to design next-generation combustion engines that run on these fuels. To achieve these objectives, a combined experimental, modeling and computational study is being carried out.

Background

The transportation sector contributes approximately one-third of the total greenhouse gas (GHG) emissions in the United States. Reduction of these emissions in any significant amount will have important impact on the total environmental GHG load.

Various pathways, including improvements in the efficiency of overall vehicle/fuel systems, such as hybrid and new high-efficiency diesel engines, and use of alternative fuels to replace or supplement petroleum-based fuels, have been proposed to achieve these reductions.

A strategy which addresses both of these pathways is the use of oxygenated compounds for fuels. There are a variety of possible feedstocks that can be used for synthesizing these oxygenates, including petroleum, oil sands, Fisher-Tropsch liquids, biomass, and algae, with the last two of these being approximately carbon neutral for the environment. These oxygenates are also especially attractive for use in the next generation of advanced Diesel engines and diesel-hybrids because of the inherently high thermal efficiencies of these engines compared to spark ignition engines and because the molecular structure of oxygenates results in high cetane numbers. In addition, these oxygenates offer significant potential for reduction in particulate (soot) and NOx emissions from diesel engines. The current strict regulations on these emissions have proved to be an impediment to a broader introduction of Diesel engines into the automotive and light truck sector in the United States. Hence, the use of oxygenates as fuels offers the prospects of reductions in GHG emissions owing to efficiency gains from advanced (i.e., low- or non-sooting) Diesel engine concepts.

Critical to the development of new fuels (and the continued use of current fuels) is concern about the effects of ultra-fine soot particles and associated volatile components. There are significant epidemiological and toxicological studies that link cardio-respiratory health effects with exposures to ultrafine soot particles (diameter < $0.1 \mu m$). The primary sources of these particles are motor vehicles in urban environments. Current soot mitigation methods remove large non-volatile soot particles, but not the precursors of the smaller semi-volatile particles which potentially can have large health risks. The development of cleaner, non-sooting fuels and an understanding of the chemical mechanisms which control this non-sooting behavior are urgently needed to address these risks.

Results

Approach

To meet program objectives, an experimental study of the combustion and emissions characteristics of oxygenated hydrocarbon fuel molecules is being carried out. An initial study focused on determining in a systematic way the impact of oxygen content and functionality on sooting tendencies of a variety of oxygenates to guide the selection of compounds to be studied in detail. The detailed studies are being conducted in two experimental facilities that have the capability of accessing conditions (temperature and pressure) relevant to Diesel engine combustion: a high-pressure shock tube (HPST) and a high-pressure flow reactor (HPFR). These two facilities are complementary in that they can access overlapping regimes of temperature and pressure, but provide different experimental data.

Concurrent with the experimental studies, detailed chemical models have been assembled to establish the role of fuel structure in ignition and NO_x and soot formation for a range of temperatures and pressures relevant to Diesel engine combustion.

To extrapolate the fundamental data from the experimental and modeling studies to engines, it is necessary to consider the complex combustion environment of a Diesel engine, which is governed by liquid fuel injection and evaporation, swirling flow, complex geometry and the presence of walls. Furthermore, it is necessary to consider a wide range of operating conditions in any optimization process. Hence, the detailed chemical models from the above studies will be applied in three-dimensional numerical simulations of diesel engine combustion using state-of-the-art methods that allow for the consideration of detailed chemistry. The combination of experiments and simulation will allow us to study and understand how oxygenated fuels reduce soot emissions in Diesel engines, both at a general and at a very detailed level that will guide the design of optimized fuel structures.

Shock Tube Studies

This research is based on an experimental program using shock tubes and laser diagnostic methods, combined with modeling using detailed reaction modeling. Work has advanced on five fronts: DME ignition kinetics, soot measurements in rich oxygenate combustion, decomposition pathways for methyl esters, a survey of oxygenate group ignition delay times, and ignition measurements of large oxygenates. Progress in these studies has lead to new information about the chemical mechanisms of oxygenates, new evidence for non-sooting fuels, and the first measurements of large oxygenate ignition properties.

DME Ignition Kinetics

DME is a simple ether for which several detailed reaction mechanisms are available. These mechanisms in general predict the ignition delay times for DME oxidation quite successfully. However when these mechanisms are used to predict the OH concentration time-histories during oxidation, they fail consistently. Because the concentration time-history of OH is intimately connected to the small radical pool (H- and O-atoms, OH and HO_2) and it is this radical pool that strongly affects ignition delay time, this failure is a serious concern.

Ignition delay times and OH concentration time-histories were measured in DME/O₂/Ar mixtures behind reflected shock waves for temperatures from 1175 - 1900 K, pressures from 1.6 - 6.6 bar, and equivalence ratios from 0.5 - 3.0. Ignition delay times were measured by collecting OH* emission near 307 nm, while OH time-histories were measured using laser absorption of the R₁(5) line of the A-X(0,0) transition at 306.7 nm. The ignition delay times extended the experimental database of DME to a greater range of equivalence ratios and pressures. Measured ignition delay times and OH time-histories were compared to simulations based on DME oxidation mechanisms by Fischer et al. (2000) and Zhao et al. (2007). Both mechanisms predict the magnitude of ignition delay times well. However, neither mechanism agrees with the measured OH time-histories. OH sensitivity analysis was applied and the reactions DME \leftrightarrow CH₃O + CH₃ and H + O₂ \leftrightarrow OH + O were found to be most important. Previous measurements of

DME \leftrightarrow CH₃O + CH₃ are not available above 1220 K, so the rate was directly measured in this work using the OH diagnostic. This rate accurately describes a broad range of experimental work at temperatures from 680 – 1750 K. When this rate is used in both the Fischer et al. and Zhao et al. mechanisms, agreement between measured OH and the model predictions is significantly improved at all temperatures.

Soot Measurements in Rich Oxygenate Combustion

There is a strong interest in blending Diesel fuels with oxygenates to reduce particulate emissions and with renewable fuels to effectively reduce the carbon content. However, the effect on soot yields by oxygenate addition to Diesel fuels has not been quantified. Without this soot yield quantification, the validation of Diesel engine simulation models cannot be assessed.

To investigate the influence of oxygenates on Diesel fuel combustion, soot formation in rich n-heptane/oxygen mixtures with dimethyl ether (DME), acetone, butanal, and 3pentanone additives was investigated behind reflected shock waves at pressures of 18 and 30 atm and temperatures from 1600 to 1900 K. At these conditions, soot formation histories up to 2 milliseconds were observed by simultaneously measuring the sootinduced laser light extinction at 633 nm and the light emitted by soot particles at 670 nm. Large soot yields were seen in the fuel-rich neat n-heptane experiments, as expected (although few quantitative data of this type are available in the scientific literature). It was also found that the soot formation process in these mixtures was very sensitive to temperature, and that there was significant reduction in overall soot yield in the heptane system with the addition of small quantities of oxygenates.

The reduction in soot yield in n-heptane/oxygenate mixtures led to further the investigations. When soot yields were measured with neat oxygenate-air mixtures, the soot yields were significantly reduced in the case of ketones (acetone and 3-pentanone), and were found to be zero (non-measurable) in the case of the one aldehyde (butanal, or butyraldehyde) and the one ester tested (methyl butanoate). We also observed, to our surprise, that the temperature window for soot formation was reduced to less than 50°K, which may have important implications for new engine strategies to eliminate soot.

This dramatically lower soot yield from these first few oxygenates, when tested neat rather than as additives, and the unexpectedly narrower soot-formation-temperature window, were not predicted from previous experiments or simulations. The consequences of this near-zero soot formation on the potential formulation of oxygenated hydrocarbon fuels, which could be blended from new feedstocks of oxygenates to achieve non-sooting emissions, needs to be investigated. These results also provide motivation to assess the concept of intelligent blending of new oxygenates from non-petroleum sources to simultaneously reduce CO_2 emissions while controlling combustion properties such as ignition delay.

Decomposition Pathways for Methyl Esters

Simple methyl esters, such as methyl butanoate, are studied because they are expected to decompose similarly to larger methyl esters found in bio-fuels. Accurate knowledge of

the methyl ester decomposition pathways is important, because it is the decomposition products of these fuels that determine the intermediate radical populations and it is these intermediate radical populations that directly affect the ignition properties and the sooting propensity of the methyl ester fuels. Current engine performance simulations using biofuels are based on detailed reaction mechanisms for methyl butanoate (and recently, methyl decanoate). Again as in the case with DME, while these mechanisms have been validated successfully against some experimental targets, the predicted levels of critical decomposition pathway products (e.g., CO_2) are significantly below that of our recent measurements. These new findings imply that existing bio-diesel fuel models, which rely on the rapid formation of two oxygenate radicals from methyl esters (rather than a single non-reactive CO_2 molecule) to account for the tendency for soot reduction, may have to be revisited.

The high temperature decomposition of three simple methyl esters: methyl acetate, methyl propionate and methyl butanoate, were studied behind reflected shock waves using tunable diode laser absorption of CO₂ near 2.7 μ m. CO₂ yield measurements were made over a range of temperatures of 1260-1653 K, pressures of 1.4-1.7 atm and reactant concentrations of 2-3 %, with the balance Ar. The CO₂ absorption strengths near 2.7 μ m are approximately 50 to 1000 times stronger than the bands near 2.0 μ m and 1.55 μ m, respectively, and offer opportunities for significantly more sensitive and accurate combustion measurements than previous absorption work using CO₂ bands at shorter wavelength. The experiments provide the first laser-based time-history measurements of the CO₂ yields during pyrolysis of these bio-diesel surrogate fuels in a shock tube. Model predictions for CO₂ yields during methyl butanoate pyrolysis at high temperatures, using the detailed reaction mechanisms of Fisher et al. (2000) and others, are significantly lower than those measured in this study. However, an improved methyl butanoate model which extends the recent theoretical work of Huynh and Violi (2007) provides substantially improved predictions of CO₂ yields during methyl butanoate pyrolysis.

Ignition Delay Time Measurements of Large Oxygenates

While fundamental chemical kinetic studies are more tenable with smaller molecules, practical fuels have significantly larger molecular weights. Using a new aerosol shock tube developed at Stanford University, we have taken the first vapor-phase ignition delay time measurements of methyl decanoate (MW = 186.29 g/mole).

Very recently, we have performed the first ignition delay time measurements (to our knowledge) for methyl decanoate. These measurements were made possible by the development of the new Aerosol Shock Tube in our laboratory. This facility enables high-temperature chemical measurements of low-vapor-pressure fuels without the problems (i.e., fuel cracking, low temperature oxidation and non-uniform mixtures) associated with standard methods of handling these fuels (i.e., using fuel pre-heating or high pressure injectors to load the facility.) Methyl decanoate (C₉H₁₉COOCH₃) is a long chain methyl ester with a boiling point of 224 °C. This is a chemically very complex molecule that we believe should mimic many of the important properties of even larger methyl ester oxygenates, especially those that are bio-derived. The demonstration of the capability to study large oxygenate species allows us to develop an experimental plan to

validate, for these larger oxygenates, the conclusions gained from studying smaller and more easily handled (in the laboratory) oxygenates and their functional groups.

Survey of Ignition Delay Times for Oxygenates

While there have been individual studies of some oxygenate compounds spread throughout the literature, there has not been a systematic study of the ignition properties of oxygenate fuels with a concern for fuel-to-fuel consistency and uniform conditions. Such a survey is needed to understand the differences, both large and small, between the ignition properties of alkanes (a major component of distillate fuels) and their related alkyl oxygenate derivatives.

Flow Reactor DME Oxidation Experiments

Experiments on DME oxidation also were conducted in a high-pressure flow reactor. These experiments complement the shock tube experiments, described above, in that they provide concentration profiles of stable reactant, product and intermediate species. The experimental data and profiles calculated using the Fisher et al. (2000) mechanism are in relatively good agreement.

CFD Code Development

The application of the Large Eddy Simulation (LES) technique to reactive internal combustion engine flows is a major challenge. In spite of the enormity of this task invaluable knowledge will be gained from an accurate LES study of reactive internal combustion (IC) engine flows. In LES, large-scale flow features are resolved or are directly computed, while small-scale flow features are modeled. This technique suits IC engines in a sense that the large-scale features of these flows are very inhomogeneous on account of the complex engine geometry. The inherent transient nature of LES flow field solutions is also very useful in studies of the time dependent behavior of IC engines. To perform LES studies of IC engine flows, we are developing a computational algorithm based on a structured grid solver in combination with the Immersed Boundary (IB) method. In the following, the reasons for these choices will be discussed and some test cases used to validate these tools will briefly be described.

Resolution of the highly unsteady and inhomogeneous flow in an IC engine requires both a highly accurate numerical method and a very fine mesh. In addition to being accurate, numerical method candidates must be robust enough to handle high Reynolds number turbulent flows. The two broad classes of algorithms, which are popular in the numerical simulation community, are the upwind-based schemes and the centered schemes. The approach of upwinding has the major disadvantage of introducing extra dissipation, which is not conducive to accurate simulation of turbulence. On the other hand, centered schemes are prone to oscillations and it has been shown in the last few years that discrete kinetic energy conservation is a very important property that these numerical schemes should satisfy to ensure stability [Morinishi et al., 2004; Desjardin et al., 2008]. The design of kinetic energy conserving structured grid algorithms has been advanced to a point where major questions regarding their implementation have been answered. The structured staggered schemes have the advantage of being discretely kinetic energy conserving, very accurate, and they lead to a solver that is very fast.

One of the obvious disadvantages of a structured grid solver as compared to an unstructured grid solver is its inability to represent complex geometrical features. A technique developed to allow a structured grid solver to resolve complex geometries is the Immersed Boundary (IB) method [Mittal and Iaccarino, 2005]. The basic idea behind the IB technique is that a structured grid can be superimposed over the complex geometric shape that needs to be represented. To correctly apply boundary conditions that account for the complex geometric shape, the flow solver algorithm needs to be modified in the region surrounding the complex shape. One of the major disadvantages of an unstructured grid solver or body conforming structured grid solver is the highly cumbersome and time consuming process of unstructured grid generation. Using the IB technique, this time consuming step is completely sidestepped and the method thus shortens the pre-processing time. The purpose here is to develop spatially hig-orderaccurate finite difference schemes that use the IB technique to represent the complex geometries, for example, the valves and piston bowl geometry. This has the additional advantage that the motion of the valves does not lead to a modification of the mesh. Since the valve motion is complex, moving meshes associated with this motion are often of poor quality. The piston motion, which simply results in a linear motion of the mesh points in one direction, is treated here with a moving mesh technique.

The moving mesh technique is based on the Arbitrary Lagrangian Eulerian (ALE) algorithm. In the Eulerian approach, which is usually used in fluid dynamics codes, the grid nodes are stationary. In the Lagrangian approach, often used in solid mechanics codes, the grid nodes move with the material associated with the respective grid nodes. In the ALE method, advantages of both approaches are coupled. In ALE, the grid nodes near the moving boundaries move with the same velocity as the boundaries, whereas the regions where large strain rate is observed, the grid nodes remain stationary as in the Eulerian approach.

In LES, the effect of unresolved scales is modeled using a subgrid-scale (SGS) model. The dynamic Smagorinsky model and the Lagrangian dynamic model [Meneveau et al, 2000], which have shown good potential for inhomogeneous flows, have been implemented in the code. Highly accurate combustion models for both premixed and non-premixed turbulent combustion are being used in this code. Models for oxygenates developed in the present work, will be implemented in the code. Models for spray breakup and evaporation have also been implemented in this solver. Also available in this code are the state of the art soot models based on the method of moments, which have been shown to perform very accurately, and are computationally inexpensive when compared with Monte-Carlo simulations.

In the following section, results obtained for some motored simplified piston cylinder configurations using the computational methods are presented. The results from these simulations are compared against experimental data and will help motivate the choice of the computational algorithms.

Simplified Piston Cylinder Configuration

This simplified piston cylinder configuration was designed at Imperial College [Morse et al., 19798]. The inlet pipe is always kept open, so there is no effective compression and the cycle is 2-stroke. This configuration was chosen because it is representative of some of the flow effects seen in real IC engines.

The computational tool mentioned above was used to perform Large Eddy simulation (LES) of the turbulent flow in this experimental configuration. The LES subfilter stresses are described by a dynamic Smagorinsky model, and a Lagrangian averaging procedure is employed in order to improve the evaluation of the Smagorinsky constant. The simulation was performed with around 900,000 mesh points. From the simulation, mean velocity and mean velocity fluctuations were evaluated at various locations in the cylinder and at various crank angle and compared against experimental data. These statistical quantities were obtained by averaging in the azimuthal direction and were also phase-averaged over 6 cycles.



Figure 1: Contour plot of velocity magnitude at 90-degree crank angle. Solid lines indicate locations where statistics were evaluated.



Figure 2: Mean velocity profiles at different stations for 90-degree crank angle; symbols are the measurements and solid lines are LES predictions.



Figure 3: Velocity fluctuation profiles at different stations for 90-degree crank angle; symbols are the measurements and solid lines are LES predictions

From Fig. 2 it can be seen that the numerical results for the mean velocity profiles compare very well with the experimental measurements. Figure 3 shows the profiles of velocity fluctuations at the same stations as in Fig. 2. Large velocity fluctuations are caused by the shear layer at the edge of the jet and by the formation of the boundary layer close to the cylinder walls.

Steady flow bench configuration

Overall engine performance is strongly dependent on the inlet and exhaust restriction, the valve or valve and port together. A better understanding of the flow structures formed around the valve would help in efficient design of valve geometry with high coefficient of discharge and hence higher mass flow rate. Due to the highly recirculating flow, Reynolds averaged Navier-Stokes (RANS) based modeling approaches have been found inadequate in predictions of mass flow rate through the restriction at various valve seat heights. The large-scale flow features are directly resolved in the case of LES, and hence, LES will be very useful in accurate predictions of coefficient of discharge and mass flow rate through the valves.

Figure 4 shows an iso-surface of the velocity in a steady flow bench experiment of a production engine valve colored by the pressure contours obtained from the computation. This test case was intended to be a very good validation of the IB technique for representing complex geometries.



Figure 4: Computed velocity iso-surface colored by the pressure contours on a steady flow bench; isometric view and top view.

Diesel Engine Tests

Several oxygen-containing fuels were tested in a 2.5 L common rail Diesel engine at high speed under two load conditions using a low sulfur Diesel fuel as the baseline fuel (BP 15). Gaseous and particulate matter (PM) emissions and engine performance were recorded. These tests were performed in part to determine whether data on soot suppression in shock tube experiments were consistent with data from engine testing. The effect of oxygenate addition on particulate matter emissions is shown in Fig. 5.



Figure 5: Particulate emissions for various oxygenated fuels at low and high load.

While having little effect on engine performance and gaseous emissions the addition of an oxygenate, in any form, reduced the total mass of particulate generated during engine tests at either low or high load. Pentanone produced the greatest reduction in total particulate mass. Particle size data suggests that this reduction in mass was due to the decrease in large particle formation. The small particles present at low load appear to be mostly part of the VOF. At high load these volatile compounds were present to a lesser extent in the particulate matter, possibly due to the higher exhaust temperatures. At low load, in-cylinder conditions restrict fuel pyrolysis and therefore mute the sooting tendency of the fuel. At high load the diffusion burn is dominant, allowing fuel chemistry, or the sooting tendency of the fuel, to become more of a factor.

Progress

Our experimental studies of effectiveness of various oxygenates in reducing sooting tendency of Diesel fuels, has revealed the types of oxygen moieties that are most effective in reducing particulate emissions and has shown that the effectiveness of the moiety is independent of the molecule in which the groups appear. From all oxygenated groups, the carbonyl group consistently was found to be the most efficient. Ester and ether groups were also effective in reducing soot formation, but less than the carbonyl group.

Significant progress has been made in developing a simulation capability for Diesel engine combustion and for soot formation in these engines.

Future Plans

We will perform shock tube and flow reactor experiments to determine the effects of oxygenates with different molecular structure on ignition characteristics and soot formation under Diesel engine combustion conditions. Concurrently, we will develop detailed reaction mechanisms that describe ignition and soot formation. These new experiments will focus on pure oxygenates as these have been found to be the most effective in reducing soot formation. The development of mechanisms for these oxygenates will provide calibration targets for the development and refinement of larger oxygenate species mechanisms.

The CFD modeling effort, in support of the experiments described above, will continue. Immediate objectives included coupling an unsteady flamelet chemistry solver with the LES code to provide a more accurate description of the in-cylinder chemistry and to perform realistic engine simulations with spray and combustion.

Publications

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