Large eddy simulations of diesel-fuel injection and auto-ignition at transcritical conditions

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Abstract
Large eddy simulations of transcritical injection and auto-ignition of \( n \)-dodecane in a combustion chamber are performed. To this end, a diffuse-interface method is employed that solves the compressible multi-species conservation equations, and a cubic state equation together with real-fluid transport properties is employed to describe the transcritical fluid state. The reaction chemistry is represented by a finite-rate chemistry model involving a 33-species reduced mechanism for \( n \)-dodecane. Compared to commonly employed two-phase approaches, the method presented in this work does not introduce tunable parameters for spray-breakup. Large eddy simulation calculations are performed by considering the Spray A single-hole injector at non-reacting and reacting conditions at a pressure of 60 bar and temperatures between 800 and 1200 K. Quantitative comparisons with measurements for liquid and vapor penetration lengths are performed for non-reacting conditions, and sensitivity to threshold values on mixture fraction are examined. The analysis of reacting flow simulations focuses on comparisons of the instantaneous temperature and species fields for OH and CH\textsubscript{2}O at 800 and 900 K, respectively. Quantitative comparisons with measurements for ignition delay and lift-off heights as a function of ambient temperature are performed. To examine the transient ignition phase, comparisons of radially integrated OH profiles obtained from the simulations with reported measurements for OH* are performed, showing good agreement. These results show that the large eddy simulation modeling framework adequately reproduces the corresponding ignition processes, which are relevant to realistic diesel-fuel injection systems.

Keywords
Diesel engine, transcritical injection, auto-ignition, diffuse-interface method, finite-rate chemistry

Introduction
Achieving further improvements in thermal efficiencies of reciprocating engines requires the operation at higher pressures. At these conditions, the fluid undergoes complex thermophysical processes, involving compressible fuel injection, phase transition, mixing, and heating, followed by ignition and combustion. These processes are often subject to pressure conditions exceeding the critical state. For diesel engines, the fuel is commonly injected at subcritical temperatures \( (T < T_c) \) into a mixture that undergoes a thermodynamic transition to a supercritical regime (see Figure 1 for \( n \)-dodecane). During the transient injection process, the fuel mixture traverses the Widom line.\textsuperscript{1} This transition is characterized by changes in the thermodynamic response functions.

Long-distance microscopy measurements performed by Manin et al.\textsuperscript{2} showed that the interfacial behavior of \( n \)-dodecane spray exhibits properties markedly different from classical two-phase breakup, including diminishing effects of surface tension and heat of vaporization. Dahms and colleagues\textsuperscript{3,4} employed linear gradient theory to describe conditions at which a multi-component fluid mixture transitions from two-phase breakup to single-phase mixing in a manner consistent with experimental observations. Results from this theory suggest that the transition to a supercritical state is a result of thermal gradients within the interfacial region, thereby broadening the interface and reducing the molecular

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A compressible Eulerian formulation was employed by Knudsen et al.\textsuperscript{9} to model the liquid fuel injection process under consideration of the internal nozzle flow. Mathies and Hickel\textsuperscript{10} used a conservative diffuse-interface method in which the phase separation was considered through vapor–liquid equilibrium calculations.

Two-phase reacting flow models have been employed by considering Lagrangian droplet methods and the gas-phase chemistry is described through flamelet models or transported probability density function (TPDF) methods. Pei et al.\textsuperscript{11} utilized a TPDF method to simulate multi-injection realizations, showing that the first ignition is initiated in a lean mixture and subsequently propagates to rich conditions. Wehrfritz et al.\textsuperscript{12} performed LES calculations with a flamelet generated manifold combustion model to investigate the early flame development at conditions similar to the Spray A operating point. They reported multi-stage ignition and observed formaldehyde formation prior to ignition at the tip of the fuel-rich gas jet. More recently, a coupled LES with tabulated flamelet model was employed by Kundu et al.\textsuperscript{13} to study the flame structure and ignition dynamics in the temperature range between 750 and 1100 K. Appreciable differences in flame structure were found at low-temperature conditions. Dahms et al.\textsuperscript{14} employed an unsteady flamelet calculation with detailed chemistry to describe the turbulent ignition process in high-pressure spray flames.

The objective of this study is to perform LES calculations to investigate fuel injection and auto-ignition in a single-hole diesel-fuel injector. To this end, a diffuse-interface method in conjunction with a finite-rate combustion model is employed. The mathematical model, consisting of governing equations, transport model, and chemical mechanism, is described in the next section. The experimental configuration and computational setup are then presented. This is followed by discussing simulation results and making comparisons with experimental data to examine the performance of the developed numerical framework. The article finishes with conclusions.

Mathematical model

Governing equations

The governing equations for the diffuse-interface method are the Favre-filtered conservation laws for mass, momentum, total energy, and species, taking the following form

\begin{align}
\partial_t \rho + \nabla \cdot (\rho \mathbf{u}) &= 0 \\
\partial_t (\rho \mathbf{u}) + \nabla \cdot (\rho \mathbf{u} \mathbf{u} + p \mathbf{I}) &= \nabla \cdot \mathbf{F}_r + \mathbf{f} \\
\partial_t (\rho \mathbf{v}) + \nabla \cdot [\mathbf{u} (\rho \mathbf{v} + p)] &= \nabla \cdot (\mathbf{F}_s + \mathbf{u}) - \nabla \cdot \mathbf{Q}_r + \mathbf{f} \\
\partial_t (\rho \mathbf{Y}_k) + \nabla \cdot (\rho \mathbf{u} \mathbf{Y}_k) &= - \nabla \cdot \mathbf{J}_{k,r} + \mathbf{f}_k
\end{align}
where $p$ is the density; $\mathbf{u}$ is the velocity vector; $p$ is the pressure; $e$ is the specific total energy; $\tau$ is the stress tensor; $\mathbf{q}$ is the heat flux; and $Y_k$, $J_i$, and $\omega_k$ are the mass fraction, diffusion flux, and chemical source term for species $k$, respectively. The species equations are solved for $k=1, \ldots, N-1$ where $N$ is the number of species. Subscripts $v$ and $t$ denote viscous and turbulent quantities, respectively. The system is closed with a state equation, $p = p(p, T, \mathbf{Y})$.

**Thermodynamic state description**

To accurately represent properties near the critical point, the Peng–Robinson cubic state equation is employed in this work

$$
p = \frac{RT}{v-b} - \frac{a}{v^2 + 2bv - b^2} \tag{2}
$$

where $R$ is the gas constant, $v$ is the specific volume, and the coefficients $a$ and $b$, taking into consideration effects of volume displacement and intermolecular forces, are dependent on temperature and composition, and are evaluated as

$$
a = \sum_{i=1}^{N} \sum_{j=1}^{N} X_i X_j a_{ij} \tag{3a}
$$

$$
b = \sum_{i=1}^{N} X_i b_i \tag{3b}
$$

where $X_i$ is the mole fraction of species $i$. In this work, the extended corresponding states principle and the pure fluid assumption for mixtures are adopted.

Parameters $a_{ij}$ and $b_i$ are evaluated using the recommended mixing rules by Harstad et al. Procedures for evaluating thermodynamic quantities such as internal energy, specific heat capacity, and partial enthalpy evaluating thermodynamic quantities such as internal energy, specific heat capacity, and partial enthalpy are described in detail in Ma et al.

To describe the dynamic viscosity and thermal conductivity, the high-pressure correction by Chung et al. is employed. This method is known to produce oscillations in viscosity for multi-species mixtures when both positive and negative acentric factors are present for individual species. To address this issue, a mole-fraction-weighted viscosity that is evaluated from Takahashi’s high-pressure correction.

**Chemical kinetics mechanism**

Because of the high computational cost associated with the evaluation of detailed chemical mechanisms, the representation of the reaction chemistry in large-scale simulations requires dimensional reduction. In this work, a 33-species mechanism for $n$-dodecane/air combustion is employed. This mechanism was reduced from a 54-species skeletal mechanism.

Zero-dimensional auto-ignition computations were performed at a pressure of $p = 60$ bar, initial temperatures in the range of $T = 800 – 1000$ K, and equivalence ratio in the range of $\phi = 0.5 – 2$. The results are sampled to apply the level of importance criterion using the YARC reduction tool. From this, 21 species are identified to be suitable for quasi-steady-state (QSS) approximation. A comparison of the resulting skeletal mechanism with its parent mechanism is provided in Figure 2. This comparison shows that the prediction of the ignition time is in good agreement with the skeletal mechanism for the entire range of temperature and equivalence ratio conditions.

The reduced mechanism is incorporated into the CFD solver using a Cantera library interface. This library allows for the run-time specification of QSS species and linearized quasi-steady-state approximation (L-QSSA) is applied to this selection of species. The sparse linear system for L-QSSA is solved efficiently by separating the construction of the elimination tree from factorization via Eigen.

**Numerical methods and boundary conditions**

The unstructured finite-volume solver, CHARLES, is employed in this study. The convective fluxes are discretized using the sensor-based hybrid scheme with the entropy-stable flux correction technique developed by Ma et al. This scheme combines a central scheme with a second-order essentially non-oscillatory (ENO) scheme, and a density sensor is employed for adaptation. Due to strong non-linearities inherent in the real-fluid state equation, spurious pressure oscillations are generated when a fully conservative scheme is used. To eliminate these pressure oscillations, an adaptive double-flux method is employed. A Strang-splitting scheme is applied to separate the convection,
diffusion, and reaction operators. A strong stability preserving third-order Runge-Kutta (SSP-RK3) scheme is used for the temporal integration of the non-stiff operators. The stiff chemical source term is integrated using a semi-implicit Rosenbrock-Krylov (ROK4E) scheme, which is fourth-order accurate in time and has linear cost complexity with respect to the number of species. The stability of the ROK4E scheme is achieved through the approximation of the Jacobian matrix by its low-rank Krylov-subspace projection. As few as three right-hand-side evaluations are performed over four stages. Details about the development of the ROK4E scheme can be found in Wu et al.

The turbulent stresses are modeled using a Vreman sub-grid scale (SGS) model. The turbulence/chemistry interaction is accounted for using the dynamic thickened-flame model, in which the maximum thickening factor is set to a value of 4 in this study.

The mass flux of the fuel and temperature are prescribed at the injector nozzle exit using the time-dependent rate of injection as provided by the CMT virtual injection rate generator, with default input parameters recommended by the engine combustion network (ECN) for the Spray A case. A plug flow velocity profile is prescribed at the nozzle exit without synthetic turbulence. A sensitivity analysis was performed to examine the effects of inflow turbulent and spray characteristics. These parametric investigations showed that turbulence has a negligible effect on the vapor penetration length and the liquid penetration length was found to decrease slightly with increasing turbulence intensity. The outlet pressure conditions are specified at 60 bar. Adiabatic boundary conditions are applied at the walls, and all simulations are initialized with quiescent ambient conditions. The governing equations are time-advanced at a Courant–Friedrichs–Lewy (CFL) number of unity, corresponding to a typical time step of 0.6 ns. All simulations are conducted up to 1.2 ms after the injection. The simulations were performed on 2560 Intel Xeon (E5-2698 v3) processors, and one calculation (over 1.2 ms) required approximately $1.5 \times 10^8$ CPU-hours.

**Experimental configuration and computational setup**

**Spray A configuration**

This study considers the Spray A single-hole injector configuration, representing an ECN benchmark case. The injector has a nominal diameter of 90 mm and is operated with pure n-dodecane fuel at a rail pressure of $p_{\text{rail}} = 1500$ bar.

The conditions in the chamber considered in this study are illustrated in the $p-T$ diagram of Figure 1. Both non-reacting and reacting cases are studied and correspond to 0% and 15% ambient O$_2$ compositions, respectively. Liquid n-dodecane fuel is injected at a subcritical temperature of $T_{\text{inj}} = 363$ K into the chamber at a pressure of $p_n = 60$ bar. An ambient temperature of $T_a = 900$ K is considered for the non-reacting case (Spray A conditions) and effects of low-temperature chemistry on the auto-ignition are examined by considering different ambient temperatures varying between 800 and 1200 K. At these conditions, the liquid n-dodecane undergoes a transcritical transition process before auto-ignition, during which the fuel is heated and mixed with the ambient gaseous environment.

**Computational setup**

The computational domain is represented by a cylindrical geometry with a diameter of 40 mm and a length of 80 mm. The domain is discretized by a structured mesh with hexahedral elements. The mesh is locally refined in the shear-layer region near the injector and stretched in downstream and radial directions. The minimum grid spacing is 4 $\mu$m near the injector exit, which results in approximately 20 grid points across the nozzle. The maximum grid spacing downstream is less than 50 $\mu$m. The mesh resolution was selected to resolve the ignition kernel, which was estimated from one-dimensional unsteady flamelet calculations. The total mesh size is 8.7 millions. Due to computational cost, a further mesh-sensitivity analysis was not performed. The injector geometry is not included in the simulations. Boundary conditions are provided at the exit of the injector nozzle.

**Results and discussion**

**Non-reacting conditions: fuel injection and mixing**

The transcritical injection process is studied by considering non-reacting flow conditions with ambient temperature of 900 K. Results for liquid and vapor penetration lengths are presented in Figures 3 and 4, showing the sensitivity of the threshold value for mixture fraction to predictions of the penetration length. The experimental vapor and liquid penetration lengths determined from Schlieren imaging and Mie scattering are also shown for comparison. Direct comparisons with measurements show that the vapor penetration length is nearly insensitive to the threshold value, whereas stronger variations for the liquid penetration length are observed as shown in Figure 4. Specifically, it can be seen that for mixture fraction values between 0.95 and 0.4, the predicted liquid penetration length varies between 5 and 15 mm. It was pointed out in the experimental investigations that the measured liquid penetration length is sensitive to measurement method, optical setup, threshold for demarcating the liquid phase, and the actual geometry of the injector. The resolution in either the simulation or the experiment remains insufficient for fully resolving the interfacial flow near the injector nozzle, if multi-phase flows do exist under these conditions.
Radial profiles of the scalar mixing field at three axial locations downstream of the nozzle exit are compared to measurements from Rayleigh scattering in Figure 5 for three different axial locations ($x = 17.85$, 25, and 35 mm). Multiple injections in the experiments provide ensemble-averaged statistics. In the simulation, statistics of the steady period of injection are obtained by temporally averaging between 0.6 and 1.2 ms after the start of the injection. As can be seen from Figure 5, there is overall good agreement for mean mixture fraction profiles at all three locations, while the simulation predicts slightly higher RMS values compared to the experimental data. These radial profiles together with the transient results for liquid and vapor penetration lengths, presented in Figures 3 and 4, show that the diffuse-interface LES-method is capable of predicting the turbulent mixing process between fuel and surrounding environment downstream of the injector after the dense liquid fuel is fully disintegrated. In this context, it is noted that this LES formulation does not introduce tunable model coefficients so that it is expected to be applicable to other transcritical injection configurations.

**Reacting conditions: auto-ignition**

We proceed by considering reacting conditions with the specific goal of examining effects of temperature variations on the low-temperature combustion. A direct comparison of transient simulation results for two different ambient temperatures of 800 K and 900 K are shown in Figure 6. Temperature fields are presented at several injection times along with the CH$_2$O and OH fields. Flow-field results for CH$_2$O and OH are superimposed to illustrate the spatial separation associated with the first-stage and second-stage ignition conditions. It can be seen from the temperature results that the liquid fuel jet is heated by the surrounding hot environment after being injected into the combustion chamber and the occurrence of a first-stage ignition can be observed at early ignition times (e.g. see temperature fields at 600 $\mu$s for 800 K and 300 $\mu$s for 900 K), which is associated with an incremental temperature change and the formation of CH$_2$O. The lower temperature for the case with 800 K delays the ignition by a factor of two. After the second-stage ignition process (see results in Figure 6 after 600/300 $\mu$s for 800/900 K), high-temperature regions with temperatures in excess of 2000 K can be seen downstream of the combustion chamber. It can also be seen that the ignition transition extends over a shorter axial distance and wider radial region compared to the case with higher ambient temperature.

From the species results in Figure 6, it can be seen that CH$_2$O is formed initially at the radial periphery of the jet. At later times, the maximum concentration of CH$_2$O is observed in the center of the penetrating jet. The formation of OH is associated with the subsequent consumption of CH$_2$O and the high-temperature chemistry by the second-stage ignition. High concentrations of OH are found near the edges of the penetrating jet due to the relatively low scalar dissipation rate and longer residence time in these regions.

To assess the model performance in predicting intermediate species, we compare instantaneous mass fraction contours of CH$_2$O and OH at the nominal ignition delay time for the condition of 900 K. Measurements were obtained from planar laser induced fluorescence. It can be seen from Figure 7 that there is a good qualitative agreement between LES and measurements in terms of shape, magnitude, and location of the formation of CH$_2$O. Similarly, acceptable agreement for OH is observed, with a sharper representation in the simulation results.
Figures 8 and 9 show comparisons of predicted and measured ignition delay and lift-off length for different ambient temperature conditions. Following the ECN-recommendation and criteria used in previous studies, the ignition delay time in LES was defined as the time when the maximum OH mass fraction reaches 14% of the value at quasi-steady state conditions of the flame. The lift-off length is calculated using the line-of-sight OH mass fraction results by time-averaging LES results between 0.8 and 1.2 ms during the quasi-steady-state period. In Figures 8 and 9, measurements from multiple experiments are shown with error bars indicating experimental variability. As can be seen from Figures 8 and 9, good agreement is observed for both the ignition delay time and the lift-off length between LES and experiments. The LES method quantitatively captures this behavior with respect to variations in ambient temperature. About 10% underprediction in ignition delay time from the LES can be observed. This discrepancy is partially attributed to uncertainties in the chemical mechanism and the mesh resolution. Shorter ignition delay times were also predicted by Yao et al. where the same parent skeletal chemical mechanism was adopted. Previous work utilizing flamelet-based combustion models showed that the chemical mechanism has a significant effect on the prediction of the ignition delay. Further investigations are needed to examine the sensitivity of the ignition process on the chemical mechanism and mesh resolution.

Note that although the ignition delay time for the cases with ambient temperatures between 1000 K and 1100 K are shorter than those for the condition at 900 K from homogeneous reactor calculations, the observed ignition delay time for the three-dimensional injection process exhibits a monotonic behavior with respect to the ambient temperature, demonstrating the significance of turbulent mixing and the shift of the ignition delay with respect to the stoichiometry during the ignition phase.

We complement this analysis by examining the transient ignition. Following the work by Maes et al., we compute so-called \( I_{tr} \)-plots by radially integrated planar fields of OH and CH2O.
Figure 6. Auto-ignition sequence for ambient chamber temperature of 800 K (left) and 900 K (right) showing temperature (left panels) and intermediate species of CH2O and OH (right panels). Spatial units in mm.
\[ I_{\text{et},i} = \int Y_i(x, y, t) \, dy \quad \text{with} \quad i = \{\text{OH, CH}_2\text{O}\} \] (4)

Figure 10 compared the results of computed OH intensities with OH* chemiluminescence measurements performed at Sandia National Laboratory. Operating conditions presented in this figure correspond to the ambient temperature of 900 K. Due to computational cost of the finite-rate simulation, LES calculations were only conducted until 1.2 ms so that direct comparisons are restricted to the ignition and early-stage combustion. The apparent streaks in the radially integrated intensity plots from the simulation results are due to the transient ignition, with a slope proportional to the advective velocity. The illustration of intensity plots for \( \text{CH}_2\text{O} \) and OH from the simulation confirms that the formation and consumption of \( \text{CH}_2\text{O} \) and OH are axially separated with the formaldehyde formation localized to the nozzle-near region, whereas the location of the peak-OH formation is shifted further downstream.

The computed OH-intensity during auto-ignition and early-stage combustion is in good agreement with experimentally determined OH*-chemiluminescence data. Predictions for flame penetration, represented by the upper bound of the OH profiles, are comparable to the measurements, indicating that the LES model captures the initial ignition phase. Further extensions of the simulation time are desirable to examine the combustion recession and burn-out phase.

**Conclusion**

Large eddy simulations were performed to examine the injection and auto-ignition of a diesel-fuel injector at transcritical conditions. A diffuse-interface method was employed and the ignition process was described using a finite-rate chemistry model consisting of a 33-species reduced mechanism. LES calculations of the ECN single-hole Spray A configuration at non-reacting and reacting conditions were performed. Comparisons
with experiments showed good agreement for predictions of the vapor penetration lengths, and a parametric sensitivity analysis identified variations in the liquid penetration length with respect to threshold values for the mixture fraction.

Reacting flow simulations considered five ambient temperature conditions, ranging between 800 and 1200 K. It was found that the simulations accurately capture experimentally observed dependencies of the ignition delay and lift-off with respect to temperature. The spatio-temporal evolution of intermediate species was examined by comparing radially integrated profiles of OH and CH$_2$O from measurements and simulations, showing quantitatively good agreement for ignition onset and early-stage combustion transition.

These results show that this LES method in conjunction with a diffuse-interface method and finite-rate chemistry is able to capture complex injection and low-temperature ignition at transcritical conditions that are relevant for diesel-fuel injectors.

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