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Numerical Simulations of Real-Fluid Reacting Sprays at Transcritical Pressures Using Multiphase Thermodynamics

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Abstract. Transcritical fuel sprays form an indispensable part of high-pressure energy-conversion systems. Modeling the complex real-fluid effects in the high-pressure multiphase regime of such sprays accurately, especially the hybrid subcritical-to-supercritical mode of evaporation during mixing fuel and oxidizer, is essential and challenging. This paper represents a novel numerical framework for accurate and efficient simulations of transcritical sprays. The spray is modeled using a diffuse interface method with multiphase thermodynamics, which couples real-fluid state equations with vapor-liquid equilibrium (VLE) calculations to compute thermo-transport properties. A physically consistent turbulence model for large-eddy simulations (LES) is used, with combustion being modeled via real finite-rate chemistry based on the fugacity of the species. The current method is accurate and free from semi-empirical drop break-up/evaporation models. LES results for the Engine Combustion Network (ECN) Spray-A benchmark demonstrate the potential of the proposed method and its advantages over traditional approaches.

Keywords: transcritical injection · turbulent combustion · multiphase thermodynamics · vapor-liquid equilibrium

1 Introduction

We consider cases where combustion is initiated by auto-ignition during or after the injection of a cold liquid fuel in a pressurized chamber filled with a hot gas oxidizer. The chamber pressure is greater than the critical pressure of the fuel but less than the cricondenbar pressure of the fuel-oxidizer mixture. This operating pressure causes the injectant to cross the two-phase region due to mixing with the environment. Such turbulent reacting sprays at transcritical pressures have drawn significant attention due to the complexity of the physical and numerical modeling. Besides real fluid effects due to the high pressure, the possibility of coexisting saturated liquid and vapor phases further complicates the modeling of transcritical fuel sprays.

The most common approach for turbulent spray modeling is primarily based on droplet breakup/collision and evaporation in the context of the Lagrangian particle tracking (LPT) method [5]. However, experimental evidence indicates that transcritical fuel injection is a mixing-controlled process instead of being controlled by droplet

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breakup or finite-rate evaporation on droplet surfaces [12]. For this reason, transcritical injectors are also often modeled in the context of the Eulerian dense-gas (DG) method [6], which assumes a single-phase working fluid with properties modeled via a suitable (cubic, e.g.) equation of state (EOS). While the DG method can be justified for combustion systems working close to the supercritical conditions, its utilization as an accurate numerical framework for transcritical sprays is questionable, since excluding the effect of the subcritical phase separation may lead to nonphysical or ill-defined states when a part of the fluid passes the meta-stable boundaries at transcritical conditions [8].

An approach with higher fidelity, which we refer to as multiphase thermodynamics (MT) [4,9,11], is to include vapor-liquid equilibrium (VLE) calculations for the multi-component real-fluid mixture, consistent with the applied EOS. In the MT method, rapid flash calculations (phase-splitting calculations) provide information on the composition and amount of saturated vapor and liquid phases, based on which the thermo-transport properties are determined by suitable mixing rules for the multiphase co-existence regions. In the case of a single-phase mixture, the MT method provides results identical to the DG method.

This paper presents the fundamentals of MT-based large-eddy simulations (LES) of transcritical sprays. Particular attention is paid to evaluating the thermo-transport properties of the working fluid in the two-phase coexistence regions. In addition to quasi-one-dimensional examples, the performance of the LES-MT method compared to conventional methods is assessed for the ECN Spray-A benchmark experiment, where transcritical n-dodecane is injected into the pressurized combustion chamber with and without oxygen.

2 Main Research Issues

In the MT method, a multi-component mixture is considered as the working fluid for which the Navier-Stokes equations (NSE) are solved. Due to the transcritical conditions during the mixing, the working fluid might be thermodynamically unstable when it passes the two-phase boundaries. In that case, the required thermo-transport properties of the working fluid are estimated based on the output of the rapid flash calculations. While one can use the amounts of saturated vapor and liquid to determine the overall mixture's extensive thermodynamic properties, estimating the dynamic viscosity and thermal conductivity requires a structure model.

3 Methodology

We solve the reacting compressible NSE free from any ideal-gas assumption in their conservative form for a working fluid with real-fluid thermo-transport and chemical properties. The working fluid can generally be a single-phase or two-phase mixture according to its local composition and thermodynamic state. In this section, the MT-based governing equations for the modeling of the real-fluid effects of such working fluid are presented.

3.1 Multiphase Thermodynamics

The NSE are closed by suitable volumetric and caloric EOSs. Additional VLE calculations must be conducted to account for the coexistence of vapor and liquid phases. This phase split information is required for calculating pressure and temperature from the mixture's density, internal energy, and mass composition.

3.1.1 State Equations

The most popular EOS is the ideal gas (IG) law, but its applicability is limited to gaseous media at relatively high temperatures and low pressures. A cubic EOS is usually selected as a suitable compromise between accuracy, complexity, and computational costs at transcritical conditions. The volumetric EOS in the general form of cubic EOS reads

$$p = RT/(\overline{\vartheta} - b) - a/[\left(\overline{\vartheta} + \delta_1 b\right)\left(\overline{\vartheta} + \delta_2 b\right)] \quad \text{with} \quad \overline{\vartheta} = \rho/W, \tag{1}$$

where ρ and W are the density and molecular weight of the mixture, a and b account for the non-ideal behavior resulting from inter-molecule attractive and repulsive forces, and δ_1 and δ_2 are two extra calibration parameters. The molar specific internal energy of the mixture, $\overline{e}$, is computed via the corresponding caloric EOS as follows:

$$\overline{e} = \textstyle\sum_{i=1}^{N} X_i \overline{h}_i^{\circ}(T) - RT + (a - T\partial a/\partial T)/[(\delta_2 - \delta_1)\,b] \ln[(\overline{\vartheta} + \delta_1 b)/(\overline{\vartheta} + \delta_2 b)], \tag{2}$$

where X_i is the mole fraction of species i. The first two terms account for the absolute internal energy of the mixture at the actual temperature but at the standard reference pressure, and the last term accounts for the internal energy change via an isothermal thermodynamic path from the standard reference pressure to the actual one. The molar specific enthalpy of species at standard pressure, $\overline{h}_i^{\circ}$, is approximated via standard NASA polynomials.

The most widely used cubic EOSs are formulated based on two constant parameters and therefore have the intrinsic limitation of a unique and universal critical compressibility factor. This results in a systematic error for the specific volume (or density) at conditions close to the critical point. To overcome this limitation, Cismondi and Mollerup [2] proposed a three-parameter cubic EOS. This so-called RKPR EOS includes the effect of the actual critical compressibility factor in the evaluation of the third parameter. The RKPR EOS is employed for our computations, a detailed rationale is given in Ref. [4].

3.1.2 Flash Calculations

Mixing at transcritical pressure might result in the coexistence of saturated vapor and liquid phases. We, therefore, solve additional VLE equations to determine the composition and amount of each phase. The VLE equations for a mixture with N components comprise N equations for species mass conservation plus $N + 2$ equations for equality of temperature, pressure, and partial molar Gibbs free energy, $\overline{g}_i$, in liquid (L) and vapor (V) phases:

$$m_i^L + m_i^V = m_i, \quad T^L = T^V, \quad p^L = p^V, \quad \overline{g}_i^L = \overline{g}_i^V, \tag{3}$$

where m_i denotes the mass of species i in the mixture. This set of $2N+2$ equations must be supplemented with two more constraints to uniquely determine the specific volume, temperature, and composition of the liquid and vapor phases. These two constraints define the type of flash problem. As we solve the conservative form of the compressible NSE, that is, transport equations for energy and density, isoenergetic-isochoric phase-splitting calculations, also known as UV-flash calculations, must be carried out. The two additional constraints for UV-flash calculations are

$$\overline{\vartheta} = (1-\alpha)\overline{\vartheta}^L + \alpha\overline{\vartheta}^V, \quad \overline{e} = (1-\alpha)\overline{e}^L + \alpha\overline{e}^V, \tag{4}$$

with α as the vapor mole fraction. To evaluate the required specific internal energies of the liquid and vapor phases, we use Eq. (2) for each phase separately, and to compute α, we use the total mass balance rewritten as

$$\alpha = (W - W^L)/(W^V - W^L). \tag{5}$$

To solve the VLE equations, we utilize the newly introduced method of Fathi and Hickel [3] that performs UV-flash calculations very fast and robust via Newton iterations with the exact Jacobian based on an effective reduction method. We note that tabulation methods are a suitable alternative for non-reacting flows. When the number of transported species is large, however, both memory requirements and table query time grow unfavorably, while the cost of the Fathi-Hickel reduction method remains low and essentially independent of the number of species. For a comprehensive review and practical implementation guidelines, readers are referred to Ref. [3].

3.2 Multiphase Transport

According to Chung et al. [1], thermal conductivity λ and dynamic viscosity μ of dense fluids are a complex function of their temperature, specific molar volume, molecular weight, acentric factor ω, and critical point information:

$$[\mu, \lambda] = f(T, \overline{\vartheta}, W, T_c, \overline{\vartheta}_c, \omega). \tag{6}$$

For a multi-component mixture, the molecular weight, critical temperature, critical molar specific volume, and acentric factor are obtained according to the composition. It is essential to note that the underlying assumption here is that the multi-component mixture is a single (stable) phase. Therefore, it would be questionable to compute the overall dynamic viscosity and thermal conductivity of a two-phase mixture via these correlations by passing the overall composition of the working fluid in the coexisting region. To correctly estimate the nominal transport properties of the working fluid, we first compute the transport properties of the saturated vapor and liquid phases via Chung correlations separately and then combine these values with a structural model to find the overall transport properties of the mixture.

For evaluating the transport properties of two-phase materials, we assume a random spatial distribution of the vapor and liquid phases within a finite-volume cell. Using the EMT model [13], the overall thermal conductivity of the mixture can be computed from its saturated liquid and vapor phase via

$$(1-\theta)(\lambda^L - \lambda)/(\lambda^L + 2\lambda) + \theta(\lambda^V - \lambda)/(\lambda^V + 2\lambda) = 0, \tag{7}$$

in which $\theta = \alpha\overline{\vartheta}^V / [\alpha\overline{\vartheta}^V + (1-\alpha)\overline{\vartheta}^L]$ is the fraction of the volume occupied by the vapor phase. Similarly, one can compute the overall dynamic viscosity of the multi-component mixture in the two-phase region.

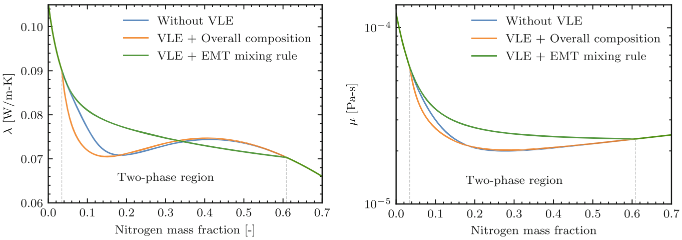

Fig. 1. Compositional functionality of the thermal conductivity (left) and dynamic viscosity (right) of n-dodecane and nitrogen mixture at 60 bar and 600 K using RKPR EOS with different inputs to Chung's method.

In Fig. 1, we compare different methods to compute the thermal conductivity and dynamic viscosity of binary mixtures of n-dodecane and nitrogen at a given temperature and pressure. All results are obtained with the RKPR EOS and Chung correlations, but with different inputs: form a dense-gas approach (no VLE calculations), from VLE calculations but using the overall (unstable in the two-phase region) composition, and from the proposed VLE-EMT model. The figure shows the proposed VLE-EMT model can reproduce transport properties free from any nonphysical oscillation of conductivity or undershooting of dynamic viscosity in the two-phase region.

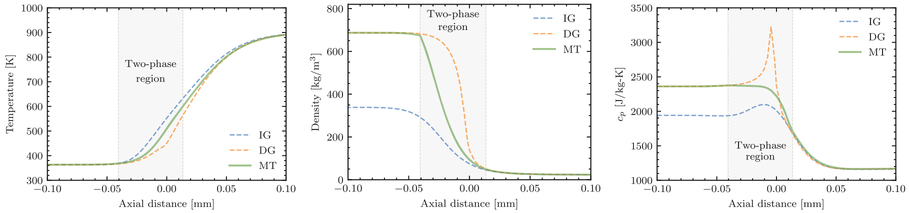

Fig. 2. The results of the MT method proposed in the current work are compared to conventional approaches of DG and GI approaches for a quasi-one-dimensional counter-flow example at ECN Spray A operating conditions.

3.3 Real-fluid Chemistry

Chemical reactions change the composition of a fluid mixture via the formation and destruction of species, which are expressed in the species mass balance equations by net mass production rates $\dot{\omega}_i$. At elevated pressures, real-fluid effects must be considered for the evaluation of this term. The species' production rates of N_r reactions can be expressed in a standard manner as

$$\dot{\omega}_i = W_i \sum_{r=1}^{N_r} (\nu_{ir}^P - \nu_{ir}^R)(k_{f,r} \prod_{j=1}^{N} C_j^{n'_{jr}} - k_{b,r} \prod_{j=1}^{N} C_j^{n''_{jr}}), \tag{8}$$

with C_j being for the concentration of species j. We use the species' fugacity for the evaluation of the concentrations $C_j \simeq f_j/(RT)$ with f_j being the fugacity of species j in the mixture. This fugacity-based evaluation of concentrations also simplifies the computation of the backward reaction coefficients via the equilibrium constant $K_\mathrm{c} = k_f/k_b$. If the reaction rates are expressed as proposed in terms of the fugacity, the equilibrium constant can be computed similarly to the ideal-gas formula:

$$K_{\mathrm{c},r} = [p^\circ/(RT)]^{\nu_r} \exp[\Delta S_r^\circ/R - \Delta H_r^\circ/(RT)], \tag{9}$$

with $\nu_r = \sum_{i=1}^{N} (\nu_{ir}^P - \nu_{ir}^R)$ being the net change in the number of species in the reaction, and ΔH_r° and ΔS_r° being the reaction enthalpy and entropy net change at the standard atmospheric pressure.

3.4 Numerical Discretization

The NSEs are discretized in space by a conservative finite-volume scheme that uses a second-order central difference method for the diffusion terms and the adaptive local deconvolution method (ALDM) of Hickel et al. [7] for the inviscid fluxes. The van Albada limiter is utilized for the mass and energy flux reconstruction to avoid spurious under- or overshoots at sharp density gradients. A third-order SSP Runge-Kutta scheme is used for advection and diffusion, and a second-order Strang-splitting scheme is used to separate the stiff chemical reactions, see Fathi et al. [4] for details. The step size is set dynamically according to the Courant-Friedrichs-Lewy stability condition.

4 Results

The transcritical condition of ECN Spray-A is selected to demonstrate the advantages of the proposed MT modeling relative to other common approaches. This spray injects liquid n-dodecane (critical pressure of 18 bar) at a low temperature of 363 K into a preheated chamber with molar composition of 15 %O_2, 6.23 %CO_2, 3.62 %H_2O, and 75.15 %N_2, temperature 900 K and pressure 60bar. ECN Spray-A has a non-cavitating nozzle, which allows us to simplify the computational setup geometry. Note that the thermodynamic model can capture in-nozzle phase change and is applicable, e.g., to the C and D injectors.

We first demonstrate the crucial role of real-fluid and multiphase effects on the transcritical mixing of the fuel and oxidizer streams using a simple quasi-one-dimensional counter-flow example without chemical reactions. For a strain rate of $500\,\mathrm{s}^{-}1$ close to the operating condition of Spray-A, Fig. 2 shows profiles of temperature, density, and heat capacity along the center line of the two opposite streams. The only difference between the DG and IG methods is the applied EOS; the RKPR EOS is used for the DG method. The MT method is based on the RKPR EOS with the aforementioned two-phase treatment. As our results indicate, the IG EOS yields an inaccurate density prediction and over-predicts the mixture's temperature. The deficiencies of the DG approach in the two-phase region manifest as a smoothed-out density solution and a non-smooth temperature profile due to the sharp nonphysical peak of isobaric heat capacity.

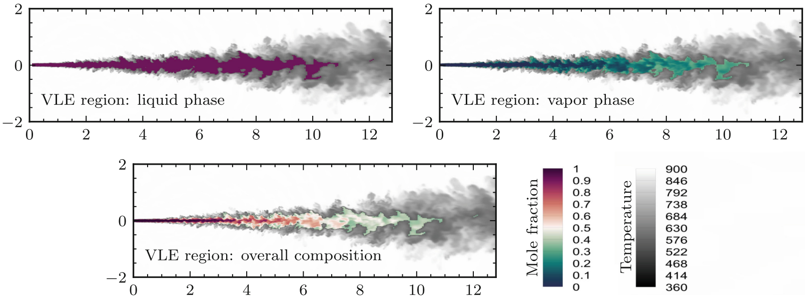

Fig. 3. LES-MT predictions of ECN Spray A. The background contour depicts the temperature field. In the two-phase region, the contours of the n-dodecane molar fraction of the saturated liquid, the saturated vapor, and the global mixture are shown.

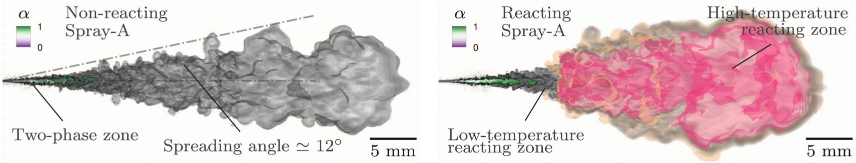

Fig. 4. LES-MT predictions of the inert (left) and reacting (right) ECN Spray A. Volume renderings at 800ţs after the start of injection.

For the LES-MT study of the ECN Spray-A, a multi-block structured grid consisting of 2864 blocks and 12.7 x 10^6 cells is generated using an adaptive mesh refinement technique. Because of the already high computational load of the multiphase thermodynamics method, a computationally efficient highly-calibrated 2-step reaction mechanism developed for the Spray-A is selected, following Hakim et al. [6].

The proposed MT method can evaluate the solubility of real-fluid mixtures at elevated pressure, as shown in Fig. 3. The figure highlights the two-phase region and shows the contours of the mole fraction of n-dodecane in the liquid, vapor, and overall mixture phases. The n-dodecane mole fraction is about 90 % in the liquid and 10 % in the vapor, which indicates the capability of the injectant to dissolve other species from the chamber gas.

Transcritical combustion typically initiates in low-temperature reacting zones, where the fluid state and transport properties deviate strongly from IG laws. Hence, making the assumption of the IG or even DG without accounting for local two-phase regions can lead to significant errors and uncertainties in transcritical combustion simulations. Volume renderings in Fig. 4 and quantitative data in Fig. 5 demonstrate that MT-LES yield accurate predictions for the spreading angle, liquid penetration lengths, vapor penetration lengths, ignition delay time, and flame lift-off length.

The LES-MT method is accurate but also has the advantage of being calibration-free. To judge its performance in relation to other approaches, we compare the vapor/liquid penetration trajectories via the LES-MT of the present work with LES-DG of Hakim et al. [6], and highly-calibrated LES-LPT of Gadalla et al. [5] with the experiments of Persson et al. [10]. Figure 5 shows results for the transient start-up interval of 0.2 ms in which we expect a similar behavior for the reacting and inert cases. The obtained results of the current LES-MT method are very close to the experimental measurements. The LES-LPT method was calibrated to match the experimental liquid length, which results in significant discrepancies in the vapor length prediction. Predictions of the DG method have the lowest accuracy in this case.

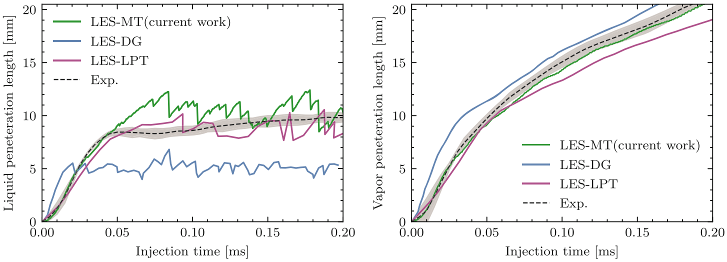

Fig. 5. Liquid and vapor penetrations computed via LES-MT in the present study [4], LES-DG of Hakim et al. [6], and highly-calibrated LES-LPT of Gadalla et al. [5] against the ECN Spray A experiment [10]

5 Conclusions and Future Work

A framework for large-eddy simulations with real-fluid multiphase thermodynamics (LES-MT) of transcritical sprays has been presented. This new methodology has been verified and validated for the transcritical ECN Spray-A at both inert and chemically reacting conditions. The predictions obtained with the proposed models are in better agreement with experimental data than results of traditional spray modeling methods from the literature.

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