VALIDATION OF AN ALL-PRESSURE FLUID DROP MODEL: HEPTANE FLUID DROPS IN NITROGEN

K. Harstad and J. Bellan
Jet Propulsion Laboratory
California Institute of Technology
Pasadena, CA 91109

Introduction

Despite the fact that supercritical fluids occur both in nature and in industrial situations, the fundamentals of their behavior is poorly understood because supercritical fluids combine the characteristics of both liquids and gases, and their behavior is not intuitive. There are several specific reasons for the lack of understanding: First, data from (mostly optical) measurements can be very misleading because regions of high density thus observed are frequently identified with liquids. A common misconception is that if in an experiment one can optically identify 'drops' and 'ligaments', the observed fluid must be in a liquid state. This inference is incorrect because in fact optical measurements detect any large change (i.e. gradients) in density. Thus, the density ratio may be well below $O(10^5)$ that characterizes its liquid/gas value, but the measurement will still identify a change in the index of refraction providing that the change is sudden (steep gradients). As shown by simulations of supercritical fluids [1], under certain conditions the density gradients may remain large during the supercritical binary fluids mixing, thus making them optically identifiable. Therefore, there is no inconsistency between the optical observation of high density regions and the fluids being in a supercritical state. A second misconception is that because a fluid has a liquid-like density, it is appropriate to model it as a liquid. However, such fluids may have liquid-like densities while their transport properties differ from those of a liquid.

Considering that the critical pressure of most fuel hydrocarbons used in Diesel and gas turbine engines is in the range of 1.5 - 3 MPa, and the fact that the maximum pressure attained in these engines is about 6 MPa, it is clear that the fuel in the combustion chamber will experience both subcritical and supercritical conditions. Studies of drop behavior over a wide range of pressures were performed in the past (Yang et al. [3], Delplanque and Sirignano [4], Haldenwang et al. [5], and the review of Givler and Abraham [6]), however none of these studies identified the crucial differences between the subcritical and supercritical behavior. In fact, in two of these studies [3], [5], it was found that the subcritical and supercritical behavior is similar as the drop diameter decreased according to the classical $d^2$-law [7] over a wide range of pressures and drop diameters.

The present study is devoted to the exploration of differences in fluid-behavior characteristics under subcritical and supercritical conditions in the particular case of heptane fluid drops in nitrogen; these substances were selected because of the availability of experimental observations for model validation.

Model equations

The configuration studied is that of a single spherical drop in a medium with specified far field conditions. These far field values are identified by the subscript 'e' and the location of the far field boundary, $R_e(t)$, is calculated in a Lagrangian way to be that of null mass flux.

The conservation equations are based upon Keizer's fluctuation theory [8] which has the distinct advantage of accounting for non-equilibrium processes. This formalism therefore leads to the most general fluid equations where the partial molar fluxes, $\mathbf{J}_i$, and the heat flux, $\mathbf{q}$, are related to thermodynamic quantities as follows:

$$\mathbf{J}_i = L_{iq} \nabla \beta - \sum_{j=1}^{N} L_{ij} \nabla (\beta \mu_j), \quad \mathbf{q} = L_{qq} \nabla \beta - \sum_{j=1}^{N} L_{qj} \nabla (\beta_j).$$

where $\beta \equiv 1/(R_u T)$, $T$ is the temperature, $R_u$ is the universal gas constant, and $\mu_j$ are the chemical potentials. Here $L_{ij}$ are the Fick's diffusion elements, $L_{qq}$ is the Fourier thermal diffusion element, $L_{iq}$ are the Soret diffusion. $L_{qj}$ are the Dufour diffusion elements, and the Onsager relations state that $L_{ij} = L_{ji}$ and $L_{iq} = L_{qi}$. Additionally, conservation of fluxes and mass in the system imply that $\sum_i m_i J_i = 0$ and $\sum_i m_i J_j = 0$ for $j \in [1,N]$ and $j = q$ where $m_i$ are the molar masses and $N$ is the total number of species.

Using the thermodynamic relationship

$$d(\beta \mu_j) = \beta (v_j dp + h_j d \ln T) + (\sum_{i=1}^{N-1} \alpha_{ij} dX_i)/X_j$$

where $v_j$ is the partial volume, $p$ is the pressure, $X_i$ is
the molar fraction, $h_j$ is the molar enthalpy, and the mass diffusion factors are

$$\alpha_{ij} = \frac{\partial X_i}{\partial \gamma_j} \frac{\partial \gamma_j}{\partial X_i} = \frac{\partial X_i}{\partial X_j} + \chi_i \frac{\partial \ln \gamma_i}{\partial X_j} \quad (3)$$

one can calculate $\overline{T}$ and $\overline{q}$ from 1 and 2. The conservation equations were derived in detail in Harstad and Bellan [1] and results were obtained for supercritical conditions. The emphasis is here on extending the calculations with the same model to the range of subcritical conditions.

**Boundary conditions**

The detailed boundary conditions at $r = R_d$ have been derived in [1]; here we describe only the new aspects that enabled the calculations to be extended to subcritical conditions. The jump conditions at the drop boundary are: mass balance; relationship between $R_d$ and the emission flux $F_{ems}$; heat balance; balance of species 1 flux; and the nonequilibrium evaporation law. Additional equations at $r = R_d$ are the momentum, and the equation of state for the mixture which is used twice (once on each side of the boundary). Thus there is a total of eight equations and nine unknowns: $\rho$, $v_b$, $X_b$, $L$, $G$, $\rho_b$, $\rho_b$, $R_d$, $T_b$, and $p_b$ and $F_{ems}$ where the subscript $b$ denotes the drop boundary, $\rho$ is the density, and superscripts $L$ and $G$ denote the inside and outside regions of the drop; variable $u_b$ is obtained by integrating the drop continuity equation starting at $r = 0$. A ninth independent relationship exists only under subcritical conditions and is related to the existence of a surface, as discussed below.

The indeterminacy of the boundary conditions for a fluid drop under supercritical conditions has already been discussed by Harstad and Bellan [1]. This is physically understandable since there is no true surface, and thus there is an arbitrariness as to the choice of the boundary to follow. At least three choices are reasonable: One may follow the pure fluid boundary as was done by Harstad and Bellan [1]. Another possibility is to follow the initial boundary separating the two fluids, this being the choice in the present calculation. The third possibility is to follow the point of maximum density gradient; although this is not the present choice, the point of maximum density gradient is calculated here a posteriori to indicate the location of the optically identified fluid drop. In contrast, under subcritical conditions the boundary to follow is the drop surface and the problem is fully determined.

There are other important consequences of the existence or lack of a surface at $r = R_d$. For example, under strong evaporative conditions a mass fraction 'film' layer exists inside the drop [9] and the thickness of this layer, $\delta r \ll \Delta r$ where $\Delta r$ is the distance from the surface to the first grid point inside the drop. A detailed analysis [2] shows that an effective mass diffusivity $D_{eff}$ can be defined with the consequence that a film layer exists when $F_{ems} \gg \rho D_{eff}/\Delta r$. The value of $D_{eff}$ and that of an equivalent thermal conductivity $\lambda_{eff}$ were calculated under the quasi-steady assumption in [2] by finding two linear combinations of $T$ and the mass fraction $Y_i$ for which the transport matrix can be approximately diagonalized. In diagonal form, the characteristic length scales for diffusional transport of these two new variables are apparent, and this allows the definition of $D_{eff}$ and $\lambda_{eff}$ [2]. Previous calculations [2] show that $\lambda_{eff} \geq \lambda$ and that $D_{eff} \leq D$. These definitions also allow the calculation of an effective Lewis number $Le_{eff} \equiv \lambda_{eff}/(nC_p D_{eff})$ once the values of the dependent variables are known. The quasi-steady assumption does not remove the generality of the estimate since the essence of the estimate is that of a characteristic length. One of the most important consequences of the mass fraction film layer exists is the direct relationship that exists between $Y(R_d - \epsilon)$ and $Y(R_d + \epsilon)$, where $\epsilon \ll \max(\delta r, \delta \rho)$; it is this relationship which provides the needed additional equation to fully determine the solution at the drop surface. This relationship can be formulated by considering the difference $\Delta Y(R_d - \epsilon) \equiv Y(R_d - \epsilon) - Y(R_d + \epsilon)$ which represents the computational grid center value at the first adjacent position to the film layer inside the drop such that $\Delta r\ll \delta r$. Similarly one may define $\Delta Y(R_d - \epsilon) \equiv T(R_d - \epsilon) - T(R_d + \epsilon)$ and $\Delta Y(R_d - \epsilon) \equiv \rho(R_d - \epsilon) - \rho(R_d + \epsilon)$. The variable $\xi_j \equiv \exp(\varphi_j - \varphi_j^0)$, where $\varphi$ is the fugacity, quantifies the $Y_j$ jump across the drop surface and can be calculated from the state equation. For example, under strict equilibrium evaporation (i.e. $F_{ems} = 0$) conditions, $\xi_j = 1$. For finite $F_{ems}$ and for a binary mixture system, its ratio to a reference state $F_{ref}(\xi_1, \xi_2)$ can be defined by

$$F_{ref} = \epsilon_F F_{ref} \quad (4)$$

where

$$F_{ref} \equiv B_1(\xi_1 - \xi_1) + B_2(\xi_2 - \xi_2) \quad (5)$$

$$B_j \equiv \alpha_{ij} m_j u_{ij} n_j, \quad j = 1, 2 \quad (6)$$

$$\xi_1 \equiv \{|B_2 \xi_2 Y(R_d - \Delta r) + B_1 \xi_1 Y(R_d - \Delta r)|/|B_2 Y_1(R_d - \Delta r) + B_1 Y_2(R_d - \Delta r)|\} \quad (7)$$

where $\alpha_{ij}$ is an accommodation coefficient, $u_{ij}$ is the mean molecular velocity crossing the plane in one direction, and $n$ is the number of moles per unit volume [1]; consistently, $F_{ref}(1, 1) = 0$. A detailed analysis of the film layer yields then $X_1^e(R_d - \epsilon), X^e_2(R_d + \epsilon), X^e_1(R_d - \epsilon)$ and $X^e_2(R_d + \epsilon)$ in terms of $\epsilon_F$ and the $\xi$'s which provides the additional relationship that allows closure of the system of equations at the drop boundary.

Since under supercritical conditions the concept of latent heat, and therefore of evaporation, is not applicable, the above analysis does not hold. However, the film layer computational approach is still necessary if $Pe_{grid} \gg O(1)$ in order to insure that all scales are
resolved. Therefore, the formalism of the film layer is retained for computational purposes even under supercritical conditions, although the layer no longer exists physically. Essentially, the solution in the supercritical regime has a diffusive character, whereas in the subcritical regime it has a diffusive-convective character where the convective part is introduced by the film layer and the evaporation.

Results

The present simulations are performed for an n-heptane drop in nitrogen because it is the set of binary substances which is best documented experimentally. The equations of state have been calculated according to the procedure described in Harstad et al. [10], and the calculation of properties has been described in Harstad and Bellan [1]. The purpose of these simulations is to validate the model.

The only data that can be used for comparisons is that obtained under evaporative rather than burning conditions, since in the last case the flame temperature that acts as the far field boundary is unknown. Furthermore, as shown below, it is only microgravity data that can be considered valid for these comparisons because normal gravity data has unavoidable convective effects that are not modeled here. Additionally, since all high pressure microgravity drop evaporation experiments were performed with suspended drops, even these data are clearly not totally equivalent to our simulation results which are obtained for a free floating drop.

To our knowledge, microgravity obtained data with $C_7H_{16}$ drops evaporating in $N_2$ were reported only by Sato [11] and Nomura et al. [12]. In their experiments the 0.7 - 1 mm drops were suspended from a fiber of at least 100μm diameter. The $C_7H_{16}$ drop evaporation experiments of Chauveau et al. [13] were conducted only in normal gravity, whereas their reported microgravity experiments were of burning drops. Therefore, our comparison focuses on the data of [11] and [12], while also considering for reference (see Table 1) the more recent normal gravity data of Morin et al. [14] for 1.15 mm drops, instead of that of Chauveau et al. [13].

The simulations were performed for nominal initial conditions matching the experimental data: $R_0^\theta = 0.35$ mm except for the comparison with Sato’s [11] data which was performed for $R_0^\theta = 0.5$ mm, and $T_{2,b}^\theta = 300$ K. The far field conditions are located at $R_0^\theta = 4$ mm where $T_c$ and $p_c$ are specified consistent with those of the experiments and $y_c^\theta = 0$. The fluid drop is initially composed of pure heptane ($T_c = 540.3$ K, $p_c = 2.76$ MPa), while the surrounding is nitrogen ($T_c = 126.2$ K, $p_c = 3.39$ MPa); in order to avoid an initial unphysical discontinuity, a minute amount of heptane exists initially in the drop surroundings, its distribution vanishing with increasing $r$. For the same reason, although the fluid drop temperature and drop-surroundings fluid composition are assumed initially uniform, a set of computational initial conditions (i.e. spatial profiles of the variables) are calculated for each simulation by satisfying the nominal initial conditions at the domain boundaries and the jump conditions at $R_d$.

In all of the discussions below, ‘subcritical’ and ‘supercritical’ qualifications will be used with respect to the heptane critical point, and not with respect to the critical point of the mixture which varies according to the local composition.

Determination of thermal diffusion factors from high temperature data

To proceed with the calculation, one must specify values for the thermal diffusion factors, $\alpha$, which can be defined either from the Irving-Kirkwood (IK) or the Bearman-Kirkwood (BK) form of the heat flux [15] and $\alpha_{BK} = \alpha_{IK} - \alpha_h$. However, values of $\alpha_{BK}$ are not well known for most substances, except at atmospheric conditions where they can be calculated from kinetic theory. Since we are here interested in calculations at considerably larger pressures, the question arises as how to calculate $\alpha_{BK}$. For this purpose, our premise is that if it can be shown that $\alpha_{IK}$ is very small, in fact it can be considered negligible with respect to $\alpha_{BK} - \alpha_{IK}$, and then $\alpha_{BK} \approx \alpha_h$ which is only a function of thermodynamic quantities [15]. Since $\alpha_h$ is calculated from thermodynamics, this would provide an approximate value for $\alpha_{BK}$ for all $(p,T)$ conditions where $\alpha_{IK}/\alpha^* = \sigma$ is very small having defined $\alpha^* = \max(p,T,X) | \alpha_h$. The purpose of these high temperature data comparisons is to explore whether our premise that $\alpha_{IK}/\alpha^*$ is very small is correct. For heptane/nitrogen $\alpha^* \approx 0.1$.

Shown on Fig. 1 are $(d/d^\theta)^2$ plots from our simulations portraying Nomura et al.’s [12] experiments at high temperature (745 K) in the pressure range of 0.1 - 2 MPa. In the numerical simulations, the location of the drop boundary is defined to be that of the maximum density gradient, for consistency with optical measurements. In agreement with well known theory [7], at 0.1 MPa the liquid/gas interface is found to be precisely that of maximum density gradient. With increasing $p$ the two locations still coincide for all simulations in the range 0.1 - 5 MPa investigated in this work, but the density gradient, although still substantial, decreases across the boundary as $p$ increases.

All but one of our simulations were conducted with $\alpha_{IK} = 0.01$; the remaining simulation was conducted with $\alpha_{BK} = 0.01$. Our results capture the 0.1 MPa data very well but display a somewhat earlier $d^2$-law behavior; it is unclear whether the non-coinciding part of the data and simulations fall within the experimental error since this error is not provided with the data. The data at 0.5 MPa is compared with results from simulations with both $\alpha_{BK}$ and $\alpha_{IK}$ specified as 0.01. It is clear that the $\alpha_{BK} = 0.01$ results fall short of agreement with the data, and in fact show a typical large increase in the evaporation time that was obtained with $\alpha_{BK} = 0.01$.
at other pressures as well. In contrast, the $\alpha_{IK} = 0.01$ results capture the nonlinear portion of the curve very well with a small discrepancy in the total evaporation time. Simulations and data at 2 MPa agree only during the initial time, after which the simulations display the expected smooth variation consistent with drop heating, whereas the data exhibit two discontinuities that can be explained only by the presence of the suspending fiber. Calculated slopes of the linear part of the curves, called the evaporation constant [7], $K$, are presented for comparison in Table 1 for the 0.1 and 0.5 MPa results obtained with $\alpha_{IK} = 0.01$. Despite the presence of the suspending fiber in the experiments, an excellent agreement exists between simulations and data. A similar comparison cannot be performed at 2 MPa since there is no evidence of linear behavior in the data.

**Confirmation of thermal diffusion factors from intermediate temperatures data**

Displayed in Fig. 2 are $p = 2$ MPa comparisons of simulation results at 655 K for various values of $\alpha_{IK}$, one simulation where $\alpha_{BK}$ instead of $\alpha_{IK}$ is prescribed, and Nomura et al.'s [12] data at 656 K. The numerical predictions are a very weak function of $\alpha_{IK}$ in the range -0.6 - 0.6 and agree remarkably well with the data during the initial heat up period of the drop. Eventually, the data shows a faster evaporation than our simulations, although the lack of error bars in the data make it impossible to evaluate the extent of the disagreement. It is also difficult to evaluate the influence of the fiber (during the experiment) on the evaporation process. However, results with $\alpha_{BK} = 0.01$ clearly overestimate both the growth of the drop during the initial heat up time and the drop evaporation time. These results are consistent with those of Fig. 1.

Additional comparisons between numerical predictions and data is portrayed in Fig. 3 where comparisons are made in the range 0.1 - 2 MPa between simulations at 655 K with $\alpha_{IK} = 0.01$, and data in the range 648 - 669 K. The initial heating time is again very well reproduced by the simulations, except that the predictions at 0.1 MPa display again an earlier $d^2$-law behavior. The evaporation time is very well reproduced at 0.1 MPa, and less well as the pressure increases. Since it is difficult to quantify the influence of the suspending fiber as the pressure increases, we can qualify this comparison as very encouraging.

Table 1 includes comparisons of $K$ for this intermediary temperature regime, and shows good to excellent agreement between data and predictions. This study indicates (see also below) that the value of $\alpha_{IK}/\alpha^*$ is indeed rather small and that $\alpha_{BK} \approx \alpha_K$ is correct. The assumption made in all calculations presented below is that the value of $\alpha_{IK}$ is the same small value determined at high temperatures regardless of the $(p, T)$ conditions, and thus that $\alpha_{BK} \approx \alpha_K$. This assumption might not be entirely valid, as in general $\alpha_{IK}$ is a function of both $p$ and $T$. This assumption and the fact that the data is from suspended drop experiments whereas our calculations are for free drops, might explain the 15-20% discrepancies (see below and Table 1) between data and results from simulations.

**Comparison with data at low temperatures.**

The low temperature data of Nomura et al.[12] and Sato [11] (Sato’s data was approximated from his figure) is shown in Fig. 4 along with our numerical predictions at 445 K, 470 K and 495 K using $\alpha_{IK} = 0.01$. The temperature range for Nomura et al.’s [12] data is 466 - 493 K whereas Sato’s [11] data was obtained at 445 K; the data in [12] is in the 0.1 - 5 MPa range, whereas that of [11] is at 2 MPa. The comparisons are very good at low $p$ and deteriorate as $p$ increases. The predictions and data [12] agree remarkably well at 0.1 and 0.5 MPa, whereas at 1 MPa the evaporation time is slightly overpredicted by the simulations. Nevertheless, the calculated and measured evaporation constant (Table 1) show very good agreement at all three pressures. The 2 MPa numerical results approximate the $d^2$ experimental variation [11] fairly well, and the agreement in the value of $K$ (Table 1) is excellent. At $p = 5$ MPa, our simulation of a free drop shows an increased heating time, whereas the suspended drop in the experiment shows a decreased heating time with respect to the 0.1 MPa case. The difference between the experimental conditions and those of the simulations explains the disagreement in the heat up time, although the rate of regression of the largest gradient location is surprisingly well predicted. Since at 5 MPa the conditions are supercritical, there is no evaporation and the concept of evaporation constant is irrelevant, although comparisons between the rates of regression are still meaningful.

**Conclusions**

A model of fluid behavior under both sub- and supercritical thermodynamic conditions has been discussed with particular emphasis on the different physics according to the initial conditions with respect to the critical point. The model has been exercised for a fluid drop for which data are available for model validation. The drop is typically colder than its surroundings whose far field conditions are prescribed. In the subcritical regime and for large emission rates from the drop, there exists a film layer in the inner part of the drop surface and the solution of the equations has a convective-diffusive character. In the supercritical regime, there is no material surface to follow, and this introduces an indeterminacy in the boundary conditions. To resolve this indeterminacy one must follow an arbitrary boundary of interest which is here that of the initial fluid drop. The solution has then a pure diffusive character, and from this solution we calculate the location of the highest density
gradient which we identify with the optically observable fluid drop.

The model was exercised for a heptane drop in nitrogen because of the existing data available for comparison. Simulations obtained with this model were validated with microgravity experimental data for large drops over a wide range of temperatures and pressures. The large temperature data were used to determine the value of the thermal diffusion factor and further validations were conducted with this fixed value. The agreement between predictions and data is excellent at atmospheric pressure and becomes fair at supercritical pressure, whereas the rate of regression of the point of maximum density gradient is remarkably well predicted at all pressures. The numerical predictions show that the traditional d²-law is obeyed only in the subcritical regime. As the pressure is increased, d² becomes non-monotonic with time, with a slope whose magnitude increases as a function of time. Thus, we initially identify a heating period during which the drop size may increase, followed by a period during which the size is continuously reduced. The duration of the heat-up period increases with far field pressure.

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References


Table 1: Maximum regression rate of the maximum density gradient location, $K$ in mm$^2$/s, obtained from the current model (ap), Nomura et al.'s microgravity experimental data (Nom), Sato's microgravity and normal gravity experimental data (Sat), and Morin et al.'s normal-gravity data (Mor). The Nomura et al.'s and Morin et al.'s data were provided by the authors, and Sato's values were read on their graph following the directions given in their paper. In the simulations $T_d^o = 300$K and $d^o = 0.7$mm, while Nomura et al.'s $d^o$ was 0.6 - 0.8mm, Sato's was 1mm, and Morin et al.'s was 1 - 1.5 mm.

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Fig. 1 High temperature comparisons. $R_d^o = 0.35$ mm; $R_e^o = 4$ mm, $Y_e^o = 0$ and $T_d^o,b = 300$ K. In the far field $T_e$ and $p_c$ are specified as in the experiments. Simulations at $T_e = 745$ K and $p_c$: 0.1MPa ---; 0.5MPa, $\alpha_{IK} = 0.01$ ---; 0.5MPa, $\alpha_{BK} = 0.01$ ---; 2MPa ---. Data: 741 K and 0.1MPa ■; 749 K and 0.5MPa ▲; 746K and 2MPa △.

Fig. 2 Intermediary temperature comparisons at 2MPa. $R_d^o = 0.35$ mm; $R_e^o = 4$ mm, $Y_e^o = 0$ and $T_d^o,b = 300$ K. Simulations at 655 K; $\alpha_{IK} = 0.01$ ---; 0.3 ---; 0.6 ---; 0.6 ---; $\alpha_{BK} = 0.01$ ---. Data at 656 K: ■.

Fig. 3 Intermediary temperature comparisons. $R_d^o = 0.35$ mm; $R_e^o = 4$ mm, $Y_e^o = 0$ and $T_d^o,b = 300$ K. Simulations at 655 K: 0.1MPa ---; 0.5MPa ---; 1MPa ---; 2MPa ---; 2MPa ---. Data: 648 K and 0.1MPa □; 669 K and 0.5MPa ▲; 669 K and 1MPa ■; 656 K and 2MPa △.

Fig. 4 Low temperature comparisons. $R_d^o = 0.35$ mm except at 445K where $R_d^o = 0.5$ mm; $R_e^o = 4$ mm, $Y_e^o = 0$ and $T_d^o,b = 300$ K. Simulations at 470 K: 0.1 MPa ---; 0.3 MPa ---; 1 MPa ---; at 445 K and 2 MPa ---; at 495 K and 5 MPa ---. Data: 471 K and 0.1 MPa □; 468 K and 0.5 MPa ▲; 466 K and 1 MPa ▼; 445 K and 2 MPa ▼; 452 K and 2MPa ▽; 493 K and 5 MPa ▲.