

Modeling Evaporation and Microexplosion of Water-in-Alkane Emulsion Droplets

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Introduction

Water is sometimes added to fossil-fuel-based combustion processes by separate injection (liquid or steam) inside the combustion chamber, or directly to the reacting fuel by means of emulsifying processes. Furthermore, the development of coal-to-liquid and liquid synfuel technologies as well as the salutary growing use of biomass pyrolysis oils as renewable energy, have also led to a substantial increase in the water fraction with respect to other components. For example, biomass pyrolysis oils have up to 25wt% water in their final composition [1].

According to Dryer (1976) [2] and, in a similar classification to Law (1977) [3] the potential benefits of adding water to the evaporation/combustion process can be classified as those arising from the chemical reactions kinetics and those arising from the so-called secondary-atomization effect or micro-explosion. Concerning chemical reactions kinetics, the resulting effects of adding water range from the passive heat sink effect to more active contributions where water vapor acts as a catalyzer on some intermediate reactions or affects the flame velocity. Moreover, independently from the type of fuel (fossil, synthetic or liquid-coal) the presence of water in such multicomponent-oil mixtures leads to a higher water vapor concentration and a temperature reduction in the gas fuel-rich region [2]. This can significantly reduce the gas phase soot formation process and diminish the chemical activities at the flame, which should reduce the production of NO_x [2]. Also, the reduced temperature can be interpreted in terms of lost of heat supply for the cracking reactions that lead to the formation of carbonaceous residue (coke) [4]. Besides thermo-chemical effects, the presence of water in forms of a non- or partially-miscible component as in water-in-alkane emulsions or water-alkane mixtures leads also to a mechanical effect, namely microexplosion, which improves the combustion efficiency.

Microexplosion occurs when the water mixed in the fuel droplet under various degrees of miscibility reaches its superheat limit [5]. Then, the onset of homogeneous nucleation in a water-fuel mixture or the onset of heterogeneous nucleation at the interface of a water microdroplet embedded in the fuel parent droplet both lead to disruptive boiling followed by secondary atomization of the parent droplet with various intensities. Due to its origin it was expected [3] and experimentally proven [6], [7], that only high-boiling point continuous phases would actually lead to micro-explosions. The benefits are a substantial increase in the surface of evaporation and an enhanced mixing of fuel vapor with air. Recent studies concerning these emerging fuels have proven that microexplosion events are not marginal and therefore can substantially contribute to the overall atomization process.

In order to account for all the aforementioned beneficial effects one has to capture accurately their respective kinetics. In particular, some of those beneficial effects depend upon the onset of microexplosion, thus upon the superheat limit of water in the fuel droplet. Therefore, we aim here the coupling of the theory of Blander and Katz (1975) [8] and, Avedisian and Andres (1978) [5] for the kinetic limit of superheat to an all-pressure emulsified-droplet evaporation model. Hereby, we shall be able to predict the onset of microexplosion. Then, following the model validation based on experimental results from the literature we study the influence of the ambient pressure as well as the emulsion characteristics on the onset of microexplosion.

Model for Water-in-Alkane Emulsion Droplet Evaporation

We consider the spherically symmetric isobaric evaporation of an emulsified fuel droplet. No chemical exothermic reactions are accounted for and the water-in-alkane emulsion droplet is in a constant atmosphere. The gas phase processes are assumed quasi-steady with respect to all liquid-phase processes. Since water is emulsified within the parent droplet and is also non-miscible with the n -alkanes of interest in the present study, both water and n -alkane vaporize independently of their relative concentrations in the liquid mixture. The water micro-droplets are large enough to consider surface tension effects unimportant at the scale of the parent droplet, thus it is reasonable to expect that the equilibrium vapor-pressure of the emulsion is the sum of the vapor pressures of n -alkane and water in their pure state. This has been verified experimentally [7].

Droplet evaporation global model (0-D)

Using the well known solution for the gas-phase convective-diffusive heat and mass transport equations in the vicinity of the droplet surface [9], we derive the following system of equations controlling the transient evolution of the droplet diameter, the species mass fractions, and the temperature of a single water-in-alkane emulsion droplet:

$$\frac{dD}{dt} = \frac{2}{\pi\rho^{(l)}D^2} \frac{dM}{dt} - \frac{D}{3\rho^{(l)}} \frac{d\rho^{(l)}}{dT} \frac{dT^{(l)}}{dt} \quad (1)$$

$$\frac{dY_\eta^{(l)}}{dt} = \frac{1}{M} \frac{dM}{dt} \left[Y_\eta^{(s)} - Y_\eta^{(l)} + \frac{Y_\eta^{(s)} - Y_\eta^{(\infty)}}{B} \right] \quad (\eta \in [\text{water, fuel}]) \quad (2)$$

$$\frac{dT^{(l)}}{dt} = \frac{6\lambda^{(s)}}{\rho^{(l)}C_p^{(l)}D^2} Nu (T^{(\infty)} - T^{(l)}) + \frac{L_v}{MC_p^{(l)}} \frac{dM}{dt} + Q_q \quad (3)$$

$$\text{with } \frac{dM}{dt} = -\frac{6M\rho^{(s)}Sh\mathcal{D}}{\rho^{(l)}D^2} \ln(1+B) \quad (4)$$

Equations (1)-(3) are solved iteratively and yield timewise evolution of the primary variables namely the droplet diameter D , the water mass fraction $Y_w^{(l)}$, the fuel mass fraction $Y_f^{(l)}$ and, the droplet temperature $T^{(l)}$ respectively. B is the so-called Spalding number or mass transfer number defined as follows for water-in-alkane emulsion droplets: $B = [(Y_w^{(s)} + Y_f^{(s)}) -$

$(Y_w^{(\infty)} + Y_f^{(\infty)})/[1 - (Y_w^{(s)} + Y_f^{(s)})]$. The Sherwood (Sh) and the Nusselt (Nu) numbers are empirically modified to take into account the convective part in the rate of mass transfer and heat transfer respectively. We use the Frössling correlations: $Sh = 2 + 0.552Re_{sl}^{1/2}Sc^{1/3}$ and $Nu = 2 + 0.552Re_{sl}^{1/2}Pr^{1/3}$, where $Re_{sl} = \rho U_{sl}D/\mu$ is the droplet Reynolds number based on the slip velocity $U_{sl} = |u_i - v_i|$ for forced convection. For natural convection we use $Sh = 2 + 0.43(GrSc)^{1/4}$ and $Nu = 2 + 0.43(GrPr)^{1/4}$ [10]. Pr and Sc are the Prandtl and Schmidt number respectively. Gr is the Grashof number calculated based on the density difference: $Gr = g\rho^2D^3\Delta\rho/(\rho\mu^2)$.

In most experiments utilizing single suspended droplets, a quartz thread is used to hold the droplet during its evaporation. In Equation (3), Q_q is the conductive heat transfer through the thread. As reported in Morin *et al* (2004) [11] the heat transferred by the fiber becomes non-negligible (6.5 to 11 % for $T^{(\infty)} \in [373; 773K]$) with respect to the heat transferred by the surrounding gas as the ambient temperature increases for quiescent droplets or droplets subjected to natural convection. As in Kadota and Hiroyasu (1976) [12], we use the following semi-empirical relation to model Q_q :

$$Q_q = \frac{\pi}{2}D_q(T^{(\infty)} - T^{(l)})\sqrt{Nu'\lambda_l\lambda_q} \quad (5)$$

where D_q is the thread diameter, $\lambda_q = 1.4$ W/mK [13] is the heat conductivity of quartz. Nu' is the Nusselt number for heat transfer from a gas to a fine cylinder and has a different expression whether the cylinder is horizontal or vertical.

Boundary conditions and thermophysical properties for all-pressure conditions

The vapor mass fractions, the temperature and, the pressure in the far-field are prescribed: $Y_{[f,w]}^{(\infty)}$, $T^{(\infty)}$ and, $p^{(\infty)}$ respectively. Following the comments of Prausnitz (1998) [14] and the numerical tests of Zhu and Aggarwal (1978) [15] we employed the Peng-Robinson equation of state (EoS) [18] to model the non-ideal behavior in the gas phase as well as the vapor-liquid phase equilibrium. When the droplet surface is in mechanical and thermodynamic equilibrium, the temperature, pressure and, fugacity of each species in the gas phase is equal to the corresponding property of the same species in the liquid phase. The equality of fugacity of species η is expressed as:

$$\Phi_\eta^v X_\eta^{(v)} = \Phi_\eta^l X_\eta^{(l)} \quad (6)$$

where Φ_η is the fugacity coefficient of the η th species, and is a function of pressure, temperature and, composition. A computationally useful expression to obtain fugacity coefficients is given by:

$$RT \ln \Phi_\eta = \int_v^\infty \left[\left(\frac{\partial P}{\partial n_\eta} \right)_{T,v,n_{k \neq \eta}} - \frac{RT}{v} \right] dv - RT \ln Z \quad (7)$$

where Z is the compressibility factor and n_k the mole number of the k th species. The Peng-Robinson cubic equation of state is then substituted into Equation (7) in order to derive the fugacity coefficient of the η th species in the liquid and the gas phase mixture [18], [19]. The vapor-liquid phase equilibrium at the droplet surface is close to a "bubble-P" type problem for

which the liquid temperature and species mole fractions are boundary conditions and the vapor mole fractions are unknowns. In this particular case the pressure is also set. An iterative method is then required to obtain the equilibrium mole fractions.

Temperature- pressure- and composition-dependent thermophysical properties are used inside the droplet and the surrounding gas. The correlations for the thermophysical and transport properties are taken from Poling *et al* (2001) [19] and are listed in Table 1.

Property	Method	Mixing rule
<u>liquid properties</u>		
density $\rho^{(l)}$	Aalto <i>et al</i> (1996)	ideal
specific heat capacity $C_p^{(l)}$	Rùzicka & Domalski (1993)	ideal
latent heat of vaporization L_v	Peng-Robinson EoS	ideal
n -alkane surface tension σ_f	Brook & Bird (1955)	
water surface tension σ_w	Vergaftik's data (1975) interpolated	
<u>Gas phase properties</u>		
density $\rho^{(v)}$	Peng-Robinson EoS	Van der Waals
specific heat capacity $C_p^{(l)}$	departure function and EoS	ideal
dynamic viscosity μ	Lucas (1980, 1981, 1983)	Lucas
thermal conductivity $\lambda^{(v)}$	Stiel & Thodos (1964)	Yorizane <i>et al</i> (1983)
diffusion coefficient \mathcal{D}	Fuller <i>et al</i> (1965, 1966, 1969)	Bird <i>et al</i>

Table 1: Thermophysical and transport properties of liquid and gas. Same correlations for n -alkane and water except when otherwise mentioned.

The nucleation rate at the water/ n -alkane interface, which is used in the microexplosion model is very sensitive to the interfacial tension of water/ n -alkane interfaces. However, as mentioned by Economou and Tsouopoulos (1997) [16] there is no accurate theoretical model based on an equation of state for the solubility of hydrocarbons in water, thus no correct model for water/hydrocarbons interfacial tension. We decided to use the experimental values of Goebel and Lunkenheimer (1997) [17] and introduced a similar temperature-dependency as the one used in the surface tension models based on the Macleod-Sugden (1963) correlation, $\sigma \propto (1 - T/T_c)^n$.

Limits to the evaporation model

Law [3], [7] proposes both a distillation limit to the evaporation model, where the liquid composition adapts instantaneously to its surface condition (rapid mixing) leading to a volatility based evaporation process and a frozen limit, where the liquid composition remains constant during the droplet lifetime. The distillation limit translates into a timewise evolution of the droplet composition during the entire droplet lifetime, $Y_\eta^{(l)}(t)$ given by Equation (2), whereas the frozen limit translates into $dY_\eta^{(l)}/dt = 0$.

Also based on physical considerations concerning emulsified fuels we propose the 'segregated distillation' limit (SD) to the evaporation model. The difference with the distillation limit applicable to multicomponent mixtures of miscible liquids only resides in the limited surface vapor pressure of the dispersed phase due to the nature of an emulsion. The distribution of water micro-droplets remains uniform and there cannot be accumulation of water at the droplet surface under the assumption of higher volatility. Moreover, we assume that the water micro-droplets diffusion timescale is very long with respect to the droplet lifetime, thus their concentration at the parent droplet surface is limited by their concentration in the parent droplet liquid core. The SD limit to the evaporation model translates into a transient composition for the continuous phase with Equation (2) used together with the vapor-liquid equilibrium relations (Equation 6) and the water surface mass fraction limited to its liquid mass fraction: $Y_w^{(s)} \leq Y_w^{(l)}$.

Fuel membrane model

In addition to the segregated distillation limit we also extended and implemented the fuel membrane model [22]. Basically, we assume a segregated distillation evaporation mode until all water micro-droplets present in a surface shell about the thickness of a micro-droplet (see Figure 1(a)) have evaporated. Then, a fuel membrane (see Figure 1(b)) forms leaving the remaining water micro-droplets trapped inside their parent droplet. On the modeling level this translates into a stop in the aqueous phase evaporation as soon as a droplet-shell evaporation criteria is fulfilled.

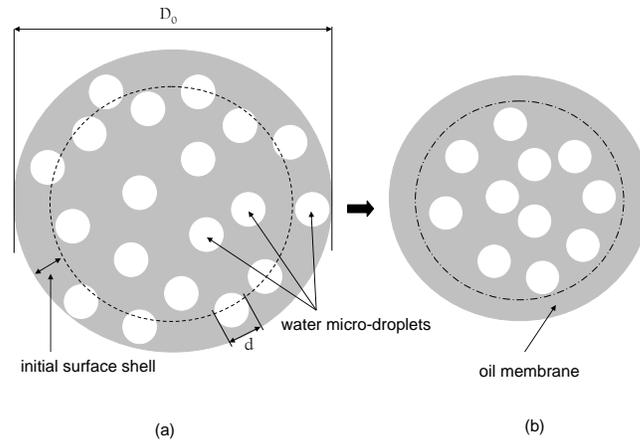


Figure 1: Sketch of the initial surface shell (a) and formation of the oil membrane (b).

The volume of water present in the initial surface shell (see Figure 1(a)) is $V_{w,0}^{(shell)} = \phi_0 V_0^{(shell)}$, where $V_0^{(shell)} = 4/3\pi(R_0^3 - (R_0 - d/2)^3)$ and R_0 the initial parent droplet radius. The end of the first stage leading to the fuel membrane formation is reached when the volume of water that has vaporized from the parent droplet is equal to $V_{w,0}^{(shell)}$. The present fuel membrane modification to the evaporation model introduces a physically expected dependency upon the water micro-droplets diameter.

Model for Microexplosion of Emulsified Droplet

The microexplosion phenomenon has its origin in the volatility difference between the water and the fuel. We have seen in the paragraph above that water is dispersed and isolated in the emulsion as relatively immobile micro-droplets. Therefore, the evaporation from the surface of an emulsified fuel droplet is essentially from the continuous fuel phase and when using high-boiling point fuels the water becomes superheated. For each emulsified fuel droplet there is a critical maximum temperature, termed its limit of superheat, at which the probability of bubble nucleation becomes appreciable. This constitutes our criteria for the onset of microexplosion. Once the energy received overcomes the minimum work required to form the nuclei the rapid vaporization of superheated water leads to the spontaneous desintegration of the parent droplet. Blander and Katz (1975) [8] and Avedisian and Andres (1978) [5] adapted the classical theory of kinetics of nucleation to the case of heterogeneous nucleation at a liquid-liquid interface. The following expression for the nucleation rate was implemented and plotted as the function of time:

$$J = \Gamma \beta_{wat} S_{wat} N \exp\left(-\frac{W}{kT}\right) \quad (8)$$

As it is more volatile than all n -alkanes under consideration, we assumed that only water evaporates into the bubble (nucleus). β_{wat} is the maximum rate of evaporation per unit surface area, N is the total number of molecules in the liquid-liquid interfacial region, Γ is the probability for a nucleus to grow to a macroscopic size, k is the Boltzmann constant and, W is the minimum work required to form a vapor bubble at a flat liquid-liquid interface. W is the summation of mechanical work, chemical work and, the work required to create the interfacial surface.

Results

Validation for pure droplet evaporation

Figure 2 (a) shows the evolution of the normalized surface area $(D/D_0)^2$ of n -heptane droplets as a function of normalized time (t/D_0^2) under atmospheric (0.1 Mpa) and moderate (0.5 Mpa; which is moderate with respect to the n -heptane critical pressure $P_c = 2.74$ Mpa) pressure conditions for different ambient temperatures. Computations [lines] performed with atmospheric pressure agree well with the experimental measurements [symbols] of Nomura *et al*(1996) [23]. Under moderate pressure conditions, the average evaporation rate, which corresponds to the slope of the linear part of the curve is in good agreement with the measurements. However, the volume expansion/heating-up period in the early stages of the droplet lifetime does not match the measurements. In order to explain such a departure between the model and the experiment we can refer to the work of Zhang (2003) [24] who performed the numerical simulation of axisymmetric n -heptane droplets evaporating in a convective high-pressure environment, solving both the liquid and the gas phase in the vicinity of the droplet.

Under initial conditions identical to the present ones, Zhang was able to show that the transients corresponding to the volume expansion/heating-up period were highly experimental-conditions-dependent and therefore, that the study of Nomura *et al* (1996) only aimed at providing experimental data for stationary droplets (average evaporation rate). In particular, by

taking into account the displacement of the droplets from the generator to the test position (a length of 60 mm in 0.16 s leading to an average velocity of 0.375 m/s) within the hot nitrogen environment, Zhang was able to reach a good agreement with respect to the measurements. In a similar fashion, we performed a second computation for each test conditions presented in Figure 2 (a) for which we modified the droplet initial temperature according to the temperature it had reached after 0.16 s under identical conditions but this time taking into account the initial convective heat transfer related to the droplet displacement. These second computations results, also displayed in Figure 2 (a) [line + symbols] agree fairly well with the measurements. This constitutes a validation of our global droplet evaporation model for atmospheric to moderate pressure conditions under microgravity conditions.

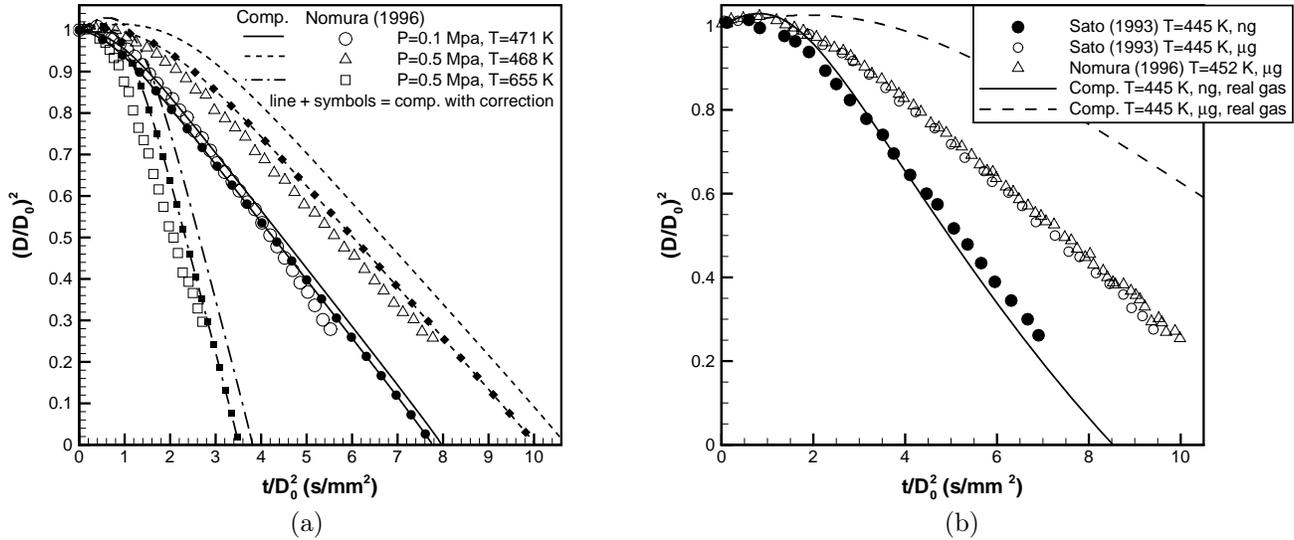


Figure 2: Evolution of the normalized droplet surface area with respect to the normalized time (t/D_0^2) for *n*-heptane single droplets. (a) atmospheric and moderate (0.5 Mpa) pressure. (b) pressure of 2 Mpa and moderate temperatures (424 K, 445 K and, 452 K).

Figure 2 (b) displays experiment and computation results for *n*-heptane droplets evaporating under subcritical pressure ($P/P_c = 0.73$) and moderate temperature conditions, and under normal gravity (ng) as well as microgravity (μg) conditions. By performing measurements under both normal gravity [filled circles] and microgravity [hollow circles] conditions Sato (1993) [25] showed that in the absence of natural convection (microgravity) the evaporation rate is smaller than under normal gravity conditions; all other boundary conditions being identical. The measurements of Nomura *et al* (1996) [hollow triangles] under microgravity are consistent with the ones of Sato (1993) [hollow circles] for identical pressures (2 Mpa) and close temperature (452 K and 445 K, respectively) conditions and, a quasi-identical experimental set-up in both studies.

One can see that the computation with natural convection [continuous line] is in very good agreement with respect to the measurements of Sato (1993) although the convective contribution to the heat and mass transfer process is modeled using semi-empirical correlations. Actually, it

is natural convection taking place under normal gravity conditions that leads to the substantial increase in the evaporation rate with respect to microgravity conditions. Hereby, we have validated our evaporation model for subcritical pressure and normal gravity conditions. Also, noteworthy the fact that the experimental measurements used for the present validation involved suspended droplets meaning a non-negligible heat conduction through the quartz thread, which was well captured by the model. Under microgravity conditions, one observes a departure of the computation with respect to the measurements which was also reported in Morin *et al* 2004 [11].

Numerical verification for the limit of superheat model and microexplosion criteria

In agreement with all experimental studies, the addition of water in a fuel droplet leads to a decrease in the overall evaporation rate (not shown) which would delay the overall evaporation duration if microexplosion did not occur. However, microexplosion events reduce considerably the overall evaporation duration. The decrease in the initial evaporation rate with respect to a non-emulsified droplets is due to the heat sink effect (higher water heat capacity with regard to n -alkanes). For microexplosion events to occur it is necessary that the parent droplet reaches the temperature of superheat and that water micro-droplets are still left by that time.

Actually, Figure 3 (a) shows that the 18% in volume water-in-hexadecane emulsion droplets computed with the frozen limit, the distillation limit or, the segregated distillation limit alone do not reach the temperature of superheat of water in hexadecane ($526K$) at atmospheric pressure. Moreover, the distillation limit leads to a complete evaporation of the water fraction in the parent droplet already at $t \simeq 0.1s$. The incrusted figure in Figure 3 (a) shows that only the segregated distillation limit coupled to the fuel membrane model allows the 18% water in volume [continuous line] and the 9% water in volume [dashed double dotted line] emulsified droplets to reach microexplosion ($T = 526 K$) as was observed experimentally [6].

The computed droplet squared diameters timewise evolutions of n -heptane and n -hexadecane droplets were compared to the experimental measurements of Wang *et al* (1984) [27] (not shown). Although the exothermic fuel-oxidation reaction was not accounted for in the computations, the comparison results demonstrated that by setting the correct far-field and initial droplet temperatures one can mimic the free-falling droplet burning experimental conditions of Wang *et al* (1984) [27]. Actually, Wang and Law (1985) [26] used the same experimental set-up to study the microexplosion process of water-in-alkane droplets.

Thus, computation results displayed in Figure 3 (b) correspond to the initial droplet and far-field temperature conditions found for the Wang *et al* (1984) test case mentioned above: $T_0^{(l)} = 480 K$ and $T^{(\infty)} = 950 K$. Plotted in Figure 3 (b) are the timewise evolution of the normalized volume and liquid temperature of a 30% in volume water-in-tetradecane emulsion droplet with $5\mu m$ water micro-droplets, evaporating under moderate pressure conditions $P=0.3$ Mpa. The kink in the droplet temperature and relative volume timewise evolution ($t \sim 0.2s$) corresponds to the time of formation of the fuel membrane. The exponential behavior of the nucleation rate defined in Equation (8), which is plotted here for values between 10000 and 20000, defines the onset of microexplosion in our model. Under these conditions, the observations of Wang and Law [26] reported a microexplosion event occurring for $(D/D_0)^3 = 0.28$. The present

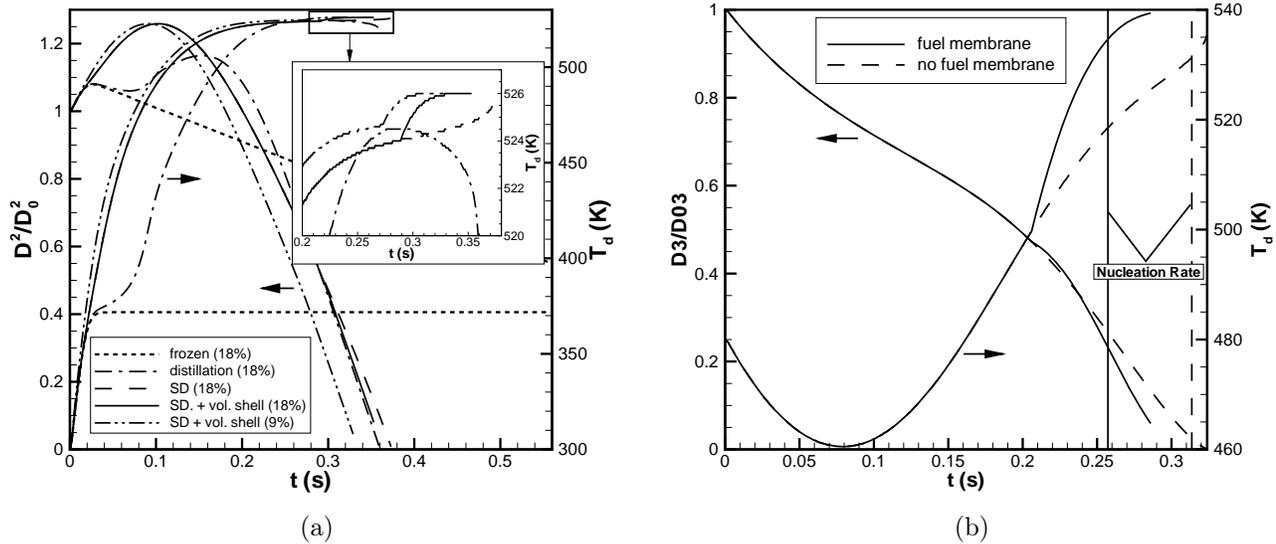


Figure 3: Computations of timewise evolution of the normalized droplet surface area. (a) water-in- n -hexadecane droplets with boundary conditions as in Lasheras *et al* (1979) [6]. Different limits to the evaporation model are tested. (b) water-in- n -tetradecane droplets as in Wang and Law (1985) [26].

model based on the SD limit plus the fuel membrane model predicts an onset for $(D/D_0)^3 = 0.23$, while the onset predicted without the fuel membrane concept is for $(D/D_0)^3 = 0.02$

The same good levels of accuracy in the prediction of the onset of microexplosion are also reached for pressures $P=0.4$ Mpa and $P=0.5$ Mpa (not shown), which were also pressure conditions used in Wang and Law [26]. Our numerical computations showed that under low pressure conditions microexplosion is enhanced by increasing the water content within the parent droplet, as was reported by Wang and Law [26]. However, this trend is reversed at higher pressures also in agreement with experimental observations [26].

Furthermore, we found that the occurrence of microexplosion is facilitated by increasing the ambient pressure. For example, the present model predicts microexplosion of a 10% water-in-hexadecane droplet evaporating in $T^{(\infty)} = 950$ K, $P=0.3$ Mpa and, $T_0^{(l)} = 500$ K for a relative volume $(D/D_0)^3 = 0.67$ (33% mass evaporated), while Wang and Law reported that 30% of the droplet volume had evaporated at the onset of microexplosion, which is in very good agreement with our computations. However, we found that no microexplosion is occurring under atmospheric pressure. This pressure dependency was also described by Wang and Law [26].

CONCLUSION

We developed an all-pressure evaporation model for water-in-alkane emulsion droplets. The heterogeneous nucleation model of Avedisian and Andres (1978) [5] was also implemented in order to model the onset of microexplosion events in all-pressure and temperature conditions.

In addition to the frozen- and distillation-like evaporation modes we introduced the segregated distillation limit. As opposed to the frozen mode, the segregated distillation approach enables the droplet to reach the superheating temperature of water in heavy n -alkanes, which is a condition necessary for the onset of micro-explosion. As opposed to the distillation limit, the segregated distillation limit ensures that water micro-droplets are still present in the parent droplet at the onset of micro-explosion. Furthermore, we improved the fuel membrane concept to capture the onset of micro-explosion events. The micro-explosion model based on a segregated distillation evaporation mode followed by the formation of a fuel membrane, and eventually the heterogeneous nucleation at the water/fuel interface is in good agreement with respect to the available experimental data.

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