Detailed simulations of droplet evaporation

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Cover: Displacement of a near–wall droplet by evaporation. Background is coloured by vapour mass fraction; droplet is coloured by temperature.

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Più che cer.co, men ri.tro.vo
Detailed simulations of droplet evaporation

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Abstract
Droplet evaporation (and condensation) is one of the most common instances of multiphase flow with phase change, encountered in nature as well as in technical and industrial applications. Examples include falling rain drops, fogs and mists, aerosol applications like electronic cigarettes and inhalation drug delivery, engineering applications like spray combustion, spray wet scrubbing or gas absorption, spray drying, flame spray pyrolysis.

Multiphase flow with phase change is a challenging topic due to the intertwined physical phenomena that govern its dynamics. Numerical simulation is an outstanding tool that enables us to gain insight in the details of the physics, often in cases when experimental studies would be too expensive, impractical or limited.

In the present work we focus on simulation of the evaporation of small droplets. We perform simulation of evaporation of a pure and two-component droplet, that includes detailed thermodynamics and variable physical and transport properties. Some of the conclusions drawn include the importance of enthalpy transport by species diffusion in the thermal budget of the system, and the identification and characterization of evaporating regimes for an azeotropic droplet.

In the second part we develop a method based on the immersed boundary concept for interface resolved numerical simulation of laminar and turbulent flows with a large number of spherical droplets that undergo evaporation or condensation.

Key words: droplet, evaporation, phase change, multicomponent, immersed boundary.
Detaljerad simulering av droppförångning

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Sammanfattning
Droppförångning (och kondensation) är en av de vanligaste fallen av flerfasflöde med fasförändring, både i naturen och i tekniska och industriella tillämpningar. Exempel är fallande regndroppar, dimma, aerosol-tillämpningar som elektroniska cigaretter och läkemedelsleverans via inandning, tekniska tillämpningar som sprayförbränning, vätskerrubbing med sprayning, gasabsorption, spraytorkning samt flammsprayspyrolys.

Flerfasflöde med fasförändring är ett utmanande ämne på grund av de sammanägda fysikaliska fenomen som styr dess dynamik. Numerisk simulering är ett utmärkt verktyg som gör det möjligt för oss att få insikt i detaljerna i fysiken, ofta i fall då experimentella studier skulle vara för dyra, opraaktiska eller begränsade.

I det nuvarande arbetet fokuserar vi på simulering av förångning av små droppar. Vi utför simulering av förångning av en ren och två–komponentdroppe, som inkluderar detaljerad termodynamik samt varierande fysikaliska och transportegenskaper. Några av de slutsatser som dras inbegriper betydelsen av entalpitransport genom diffusion av olika ämnen i systemets termiska budget samt identifiering och karakterisering av förångningsregimer för en azeotropisk droppe.

I den andra delen utvecklar vi en metod baserad på det nedsänkta rand konceptet för gränssnittskompletterad numerisk simulering av laminära och turbulenta flöden med ett stort antal sfäriska droppar som genomgår förångning eller kondensering.

Nyckelord: dropp, förångning, fasövergång, multikomponent, nedsänkt rand.
Preface

This thesis deals with detailed numerical simulation of droplet evaporation. An introduction on the basic concepts and a review of the methods are presented in the first part. The second part contains two articles. The papers are adjusted to comply with the present thesis format for consistency, but their contents have not been altered as compared with their original counterparts.


December 2017, Stockholm

Giandomenico Lupo
Division of work between authors
The main advisor for the project is Prof. Christophe Duwig (CD).

**Paper 1.** The code has been developed by Giandomenico Lupo (GL). The simulations have been performed by GL. The paper has been written by GL with feedback from CD.

**Paper 2.** The code was originally developed by Wim-Paul Breugem (WB). It has been extended by GL and Mehdi Niazi Ardekani (MN). The simulations have been performed by GL. The paper has been written by GL with feedback from CD, MN and Luca Brandt (LB).

Conferences
Part of the work in this thesis has been presented at the following international conferences. The presenting author is underlined.


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Part I

Overview and summary
Chapter 1

Introduction

Among the numerous instances of multiphase flows that surround us in nature as well as in industrial and engineering applications, flows with phase change are especially interesting for their complex interplay of physics. Three transport mechanisms (momentum, mass and energy) act simultaneously. The mass and energy distributions determine the thermodynamic state of the system, which in turn sets the conditions for the phase change. The phase change then affects the transport of all physical quantities, which in turn acts on the dynamics of the phase change itself by evolving the interphase boundary conditions.

The coupling between the different physical phenomena is sketched in Fig. 1.1. When one considers that mass and energy distributions affect the physical and transport properties as well, the feedback mechanisms become even more complex.

The different physical phenomena are associated with their own time and length scales. Numerical simulations are an invaluable tool to help untangle the interacting physics and identify the relevant scales.

The present work is concerned with a particular instance of multiphase flow with phase change: the evaporation of small droplets. This apparently
narrow phenomenon occurs in a variety of situations subject to widely different conditions: falling rain drops, fogs, aerosol applications like electronic cigarettes and inhalation drug delivery, as well as more classic industrial fields like spray combustion (Faeth 1977), spray wet scrubbing or gas absorption (Treybal 1981), and spray drying (Mezhericher et al. 2010). Flame spray pyrolysis, i.e. gas phase combustion synthesis of ceramic particles used as catalysts, pigments, etc., starting from liquid precursors, is also a novel process where droplet evaporation dynamics play an important role (Mädler et al. 2002; Strobel et al. 2006).

Many of the above mentioned applications involve spray jets. When considering the whole injection/atomization/evaporation system, we can distinguish three zones within the jet that are dominated by different phenomena (see Fig. 1.2): close to the nozzle the liquid jet breaks up into larger blobs and ligaments (primary breakup), then these further break down into smaller and smaller droplets (secondary breakup). These two regions are dominated by liquid inertia and surface tension. After secondary breakup, the flow is typically dominated by evaporation since the liquid surface area is much larger.

![Figure 1.2: Injection and atomization of a full-cone diesel spray.](image)

We focus our attention on droplet evaporation after secondary breakup, where droplets retain a spherical shape for the most part. In Chapter 1 we provide a review of droplet evaporation models and propose a new model that
includes enthalpy transport by species diffusion. We also propose a simplified model for pure droplet evaporation history and lifetime estimation. With regard to interface resolved numerical simulation, in Chapter 2 we review the most common methods that have been used in the literature to simulate spray evaporation.

In paper 1 we performed detailed numerical computation of the evaporation of an ethanol–water droplet. The topic of evaporation of non–ideal mixtures is relevant in view of the increasingly growing adoption of biofuel combustion.

In paper 2 we develop an Immersed Boundary method for interface resolved numerical simulation of flows with evaporating droplets.
Chapter 2

Models for evaporating sprays

Spray evaporation calculations embedded in general purpose flow solvers have traditionally been tackled with methods that treat the droplets as a collection of material points. The droplet dynamics within the flow are calculated by solving lagrangian equations of motion for the material points, while the evaporation dynamics are handled with integral expressions for the heat and mass fluxes originating from each droplet (Continillo & Sirignano 1990; Darabiha et al. 1993; Kee et al. 2011; Franzelli et al. 2013; Feng et al. 2015; Liñan et al. 2015).

This approach requires two main modelling endeavours: finding appropriate closed expressions for the mass and heat evaporation rates, and for the forces that drive the droplet motion.

2.1. Droplet evaporation

Classic review papers on droplet evaporation modelling are those by Faeth (1977), Law (1982), Sirignano (1983) and Aggarwal & Peng (1995), while a more recent and comprehensive review can be found in Sazhin (2006), as well as in the book by the same author (Sazhin 2014).

2.1.1. Governing equations

The physical problem involves momentum, energy and species transport in the gas phase. Common assumptions for the gas phase are incompressible flow, ideal gas, and no viscous dissipation, whereby the governing equations read:

- Momentum:
  \[ \nabla \cdot \mathbf{u} = 0; \]
  \[ \rho \frac{D\mathbf{u}}{Dt} = -\nabla p + \nabla \cdot (\mu (\nabla \mathbf{u} + \nabla \mathbf{u}^T)) . \]  
  \[ (2.1) \]

- Energy:
  \[ \rho c_p \frac{DT}{Dt} = \nabla \cdot (\kappa \nabla T) + \rho (c_{p,vap} - c_{p,inert}) D_{vap} \nabla T \cdot \nabla Y. \]
  \[ (2.2) \]

- Vapour mass fraction:
  \[ \rho \frac{DY}{Dt} = \nabla \cdot (\rho D_{vap} \nabla Y) . \]
  \[ (2.3) \]
2.1. Droplet evaporation

The mass and heat fluxes integrated over the droplet surface are:

\[ \dot{m} = \int_S (-\rho D_{vap} \nabla Y + \rho u Y) \cdot n \, dS. \]  \hspace{1cm} (2.4)

\[ \dot{q} = \int_S (-\kappa \nabla T) \cdot n \, dS. \]  \hspace{1cm} (2.5)

The enthalpy diffusion term in the gas energy equation (Eq. 2.2) arises from the different heat capacities that the vapour and inert gas carry with themselves as they diffuse into each other (Bird et al. 2007). There is no agreement in the literature as to whether this contribution to the heat transport budget should be included or can be neglected. It was included in evaporation models as early as the 1970s in the papers of Newbold & Amundson (1973) and Hubbard et al. (1975). Curtis & Farrell (1992) were able to reproduce experimental data at various pressure conditions with their model, which includes enthalpy diffusion. Harstad & Bellan (2000) use a similar approach for numerical simulation of droplet evaporation at supercritical conditions. Yang & Wong (2001), in an effort to come as close as possible to the prevalent experimental conditions in their numerical model, account for enthalpy diffusion, as well as radiation and heat conduction through the fiber on which the droplet is usually suspended during evaporation experiments. Ebrahimian & Habchi (2011) finally pinpoint the specific influence of the enthalpy diffusion contribution, showing that its exclusion leads to higher droplet temperatures and overprediction of the evaporation rate. This is supported by Lupo & Duwig (2017) (Paper 1 in the present thesis), where a model for multicomponent droplet evaporation that includes enthalpy diffusion is presented. Validation of the model is conducted by reproducing the simulations of Yang & Wong (2001), and observing again major overprediction of the evaporation rate should the enthalpy diffusion be neglected. The reason can be directly inferred from Eq. 2.2. The vapour species is being transported from the droplet surface, where it has its maximum concentration, towards regions far from the surface where its concentration is low. Heat, on the other hand, travels the opposite way, from the gas to the liquid. In short, normal gradients of temperature and vapour mass fraction have opposite sign at the surface of a droplet that is evaporating in a hot gas environment. This, in concurrence with the heat capacity of the vapour being usually bigger than that of the gas, makes the enthalpy diffusion term in Eq. 2.2 negative, thus slowing down the droplet heating and thereby the evaporation rate.

Cook (2009) shows by simple thermodynamic considerations that species diffusion without the associated enthalpy flux is an impossible scenario, as it violates the second law of thermodynamics. He then gives examples, calculated within a fully compressible framework, of the inception of anomalous temperature gradients in the flow if the enthalpy diffusion is neglected. These are more conspicuous the larger the difference in molecular weights (and therefore heat capacities) of the diffusing species is; a condition that occurs frequently in evaporation of e.g. liquid fuels. The mechanism by which these spurious
temperature gradients are suppressed, as explained by the author, is by coupling of the density field to energy through the equation of state: the diffusion of different species originates a density gradient, which can only be balanced by a velocity divergence. This divergence originates from the pressure variation caused by enthalpy diffusion. The smoothing of the density field redistributes the specific heat capacity of the mixture, suppressing the spurious temperature gradients. This particular effect is obviously not captured by an incompressible flow solver; however, in the case of evaporation, the effect on the heat equation itself seems to be large enough as not to be neglected.

Despite this fact, a large number of evaporation models actually disregard the enthalpy diffusion contribution to the heat equation (Ranz & Marshall 1952; Faeth 1977; Law 1975, 1976; Law & Law 1982; Law 1982; Renksizbulut & Yuen 1983; Tong & Sirignano 1986; Abramzon & Sirignano 1989; Yao et al. 2003; Tonini & Cossali 2012; Saha et al. 2012; Sazhin 2014; Alam et al. 2014). To the knowledge of the author, spray flow simulations with phase change have restricted themselves to a just few of these models even recently, thus overlooking the effect of enthalpy diffusion on the phase change process (Continillo & Sirignano 1990; Darabiha et al. 1993; Kee et al. 2011; Franzelli et al. 2013; Liñan et al. 2015; Feng et al. 2015; Mahiques et al. 2017).

Since the purpose of the present analysis is to derive closed expressions for the mass and energy transfer rates \( \dot{m} \) and \( \dot{q} \) pertaining to droplets that are represented by material points in the flow solver, it is assumed that the droplets are small and thus preserve spherical shape. If the surrounding gas flow is static no circulation arises in the droplet, assuming buoyancy is negligible, so that the governing equations for the liquid phase are one-dimensional and transport is only diffusive.

As the droplet radius undergoes continuous change due to the evaporation, it is convenient to define a radial coordinate \( \omega \), normalized with respect to the droplet radius \( r_s(t) \) at each instant:

\[
\omega = \frac{r}{r_s(t)}, \quad 0 \leq \omega \leq 1.
\] (2.6)

With no liquid circulation, the liquid phase is described by the energy transport equation and its global mass balance:

- **Energy**:

\[
\frac{r_s^2}{\alpha_l} \frac{\partial T_l}{\partial t} = \frac{\partial^2 T_l}{\partial \omega^2} + \left( \frac{2}{\omega} + \frac{1}{\kappa_l} \frac{\partial \kappa_l}{\partial \omega} + \frac{1}{2} \frac{\omega}{\alpha_l} \frac{\partial \alpha_l}{\partial t} \right) \frac{\partial T_l}{\partial \omega}.
\] (2.7)

- **Mass balance**:

\[
\frac{dr_s^2}{dt} = -2 \frac{\dot{m}}{4\pi r_s} + r_s^2 \int_0^1 \omega^2 \frac{\partial \rho_l}{\partial \omega} d\omega - \rho_l(\omega = 1, t) - \int_0^1 \omega^3 \frac{\partial \rho_l}{\partial \omega} d\omega.
\] (2.8)
The heat penetrating the liquid changes the droplet temperature (sensible heat) and supplies the energy for the evaporation (latent heat):

\[
\left. \frac{\partial T_l}{\partial \omega} \right|_{\omega=1} = -\frac{(\dot{q} + \dot{m} \Lambda)}{4\pi r_s \kappa_l(\omega = 1, t)}.
\] (2.9)

At the droplet surface the temperature is continuous i.e. \( T = T_l \). Moreover, thermodynamic equilibrium is the customary assumption that ties the droplet surface temperature to the vapour concentration on the gas side, providing the surface boundary condition for Eq. 2.3.

### 2.1.2. Heat and mass transfer rates

James Clerk Maxwell was perhaps the first one to provide a closure for the evaporation rate (Eq. 2.4) in 1877 (Maxwell 2011):

\[
\dot{m} = 4\pi r_s^2 \frac{D_{\text{vap}} M_{w,\text{vap}}}{RT_l} \frac{P_{\text{sat}}(T_l)}{\Delta h}.
\] (2.10)

His expression can be recovered from Eq. 2.4, assuming uniform and constant droplet temperature, a stationary gas phase, neglecting the convective term that arises from the radius regression velocity (Stefan flow), and linearizing the concentration gradient over an appropriate distance \( \Delta h \) from the surface where concentration of the vapour vanishes.

Relaxing the hypothesis of no Stefan flow, closed expressions for the evaporation rate \( \dot{m} \) and the heat transfer rate \( \dot{q} \) can be obtained if the gas phase is assumed quasi-steady and spherically symmetric, and the properties are taken as constant, or appropriately averaged. This solution was described by Fuchs (1959), albeit without considering the enthalpy diffusion term in the gas heat equation, and is at the core of virtually all evaporation models used to this day. Here we lay out a version that includes enthalpy diffusion.

We define the Spalding mass transfer number \( B_M \) for the vapour species:

\[
B_M = \frac{Y_s - Y_\infty}{1 - Y_s}; \tag{2.11}
\]

and the heat transfer number \( B_T \):

\[
B_T = \frac{1}{Le} \int_1^{1 + B_M} \zeta^{(1/Le) - 1} \exp \left[ \frac{1}{Le} \left( \frac{\bar{c}_{p,\text{vap}} - \bar{c}_{p,\text{inert}}}{\bar{c}_p} \right) (1 - Y_s) (\zeta - 1) \right] d\zeta.
\] (2.12)

Integration of Eq. 2.3 and Eq. 2.2 then yields:

\[
\dot{m} = 4\pi \bar{p} D_{\text{vap}} r_s \ln (1 + B_M); \tag{2.13}
\]

\[
\dot{q} = \frac{\dot{m} \bar{c}_p (T_s - T_\infty)}{B_T}. \tag{2.14}
\]
The average gas phase properties are evaluated at a reference temperature and composition. Hubbard et al. (1975) report some correlations for the reference states, and, noting that superior results are obtained when properties are evaluated at a state closer to the evaporating surface than the free stream, suggest a simple linear averaging:

\[ T_{\text{ref}} = T_s + A_r (T_\infty + T_s) ; \]  
\[ Y_{\text{ref}} = Y_s + A_r (Y_\infty + Y_s) ; \]

with \( A_r = 1/3 \). This rule has been prevalent among evaporation models ever since. Ebrahimian & Habchi (2011) compare the results obtained with \( A_r = 1/3 \) and \( A_r = 1/2 \) to experimental evaporation data, and show that the 1/3 rule performs significantly better at high gas temperature, while at low gas gas temperature there is hardly any difference between the two rules.

2.1.3. Evaporation in convective environment

If the gas has a net bulk velocity other than the Stefan flow, or, equivalently, if the droplet is moving through the gas, spherical symmetry is lost in the gas phase. The shear on the droplet surface also induces circulation inside the droplet, which breaks spherical symmetry in the liquid phase as well.

Nevertheless, the dominant approach for the gas phase is to introduce semiempirical correction factors for the transfer rates of a symmetric flow field, that account for effects of convection. This greatly simplifies the calculations compared to a complete solution of the three−dimensional fields in both phases.

The Nusselt and Sherwood numbers are defined respectively as nondimensional heat and mass fluxes, normalized with the diffusive fluxes in the absence of convection, so that:

\[ \dot{q} = \frac{4\pi r_s^2}{\pi} \left( T_s - T_\infty \right) ; \]
\[ \dot{m} = \frac{4\pi r_s^2}{\pi} \left( \rho D_{\text{vap}} Y_s - Y_\infty \right) . \]

If the transport is purely diffusive (i.e. the Maxwell solution), then \( Nu = Sh = 2 \). If the only mechanism of convective transport is the Stefan flow and the enthalpy diffusion term in the gas heat equation is neglected, it can be shown that:

\[ Nu = 2 \frac{\ln(1 + B_T)}{B_T} ; \]
\[ Sh = 2 \frac{\ln(1 + B_M)}{B_M(1 - Y_s)} . \]

When enthalpy diffusion is not neglected, the relation between \( B_M \) and \( B_T \) (Eq. 2.12) is not explicit anymore, and Eq. 2.18 must be replaced by:
\[ N_u = 2 \frac{\ln(1 + B_M)}{Le B_T}; \] \hspace{1cm} (2.20)

From Abramzon & Sirignano (1989) to Sazhin (2006, 2014), the factor \((1 - Y_s)\) in Eq. 2.19 is omitted in the literature, to the best of our knowledge; this is a valid approximation only when \(Y_s \ll 1\). We also note that, in the absence of enthalpy diffusion, Eq 2.12 simplifies to \(B_T = (1 + B_M)^{1/Le} - 1\), instead of \(B_T = (1 + B_M)^{\text{cp,vap}} \frac{1}{Le} - 1\) as it is erroneously reported in Abramzon & Sirignano (1989) and all the successive authors that follow their analysis \(^1\), up to Sazhin (2014).

In the case of a droplet evaporating in forced convective environment, many correlations are available in the literature. Ranz & Marshall (1952) give the following expressions, in the limit of \(B_T \to 0\) and \(B_M \to 0\) and constant droplet radius, for moderate Reynolds numbers:

\[ N_u_0 = 2 + 0.552Re^{1/2}Pr^{1/3}; \] \hspace{1cm} (2.21)
\[ Sh_0 = 2 + 0.552Re^{1/2}Sc^{1/3}. \] \hspace{1cm} (2.22)

Faeth (1977), combining the expressions of Ranz & Marshall (1952) (with a slightly different coefficient) with the leading order term in the perturbation analysis by Acrivos & Taylor (1962), valid for creeping flow (i.e. for \(Re \to 0\)), gives the following expressions for \(Re < 1800\):

\[ N_u_0 = 2 + \frac{0.555Re^{1/2}Pr^{1/3}}{\sqrt{1 + \frac{1.232}{RePr^{4/3}}}}; \] \hspace{1cm} (2.23)
\[ Sh_0 = 2 + \frac{0.555Re^{1/2}Sc^{1/3}}{\sqrt{1 + \frac{1.232}{ReSc^{4/3}}}}. \] \hspace{1cm} (2.24)

Clift et al. (1978) give the following expressions for \(Re < 400\), \(0.25 < Pr < 100\) and \(0.25 < Sc < 100\):

\[ N_u_0 = 1 + (1 + RePr)^{1/3} \max [1, Re^{0.077}] ; \] \hspace{1cm} (2.25)
\[ Sh_0 = 1 + (1 + ReSc)^{1/3} \max [1, Re^{0.077}] . \] \hspace{1cm} (2.26)

The subscript “0” on these factors indicates that they were obtained in the limit of very small evaporation flux, i.e. \(B_M = B_T = 0\). Nonetheless they were still used in many evaporation studies.

The classical way to overcome this shortcoming, and obtain expressions valid for finite values of \(B_M\) and \(B_T\), is to express the ratio of fluxes with and without finite evaporation in analogy to the case without forced convection,

\(^1\)The mistake comes from writing the gas heat equation (Eq. 2.2) with the heat capacity of the vapour instead of the heat capacity of the gas mixture for the convective term.
where the case with finite evaporation would exhibit Stefan flow, so that (Sazhin 2006):

\[
Nu = Nu_0 \frac{\ln(1 + BT)}{BT}; \quad (2.27)
\]

\[
Sh = Sh_0 \frac{\ln(1 + BM)}{BM(1 - Y_s)}. \quad (2.28)
\]

Again we note that the factor \((1 - Y_s)\) is omitted by Sazhin (2006). As before, if enthalpy diffusion in not neglected, Eq 2.27 must be replaced by:

\[
Nu = Nu_0 \frac{\ln(1 + BM)}{LeBT}. \quad (2.29)
\]

Renksizbulut & Yuen (1983), instead of Eqs. 2.27 and 2.28, give, for \(10 < Re < 150\):

\[
Nu = \left(2 + 0.57 Re^{1/2} Pr^{1/3}\right) (1 + BT)^{-0.7}; \quad (2.30)
\]

\[
Sh = \left(2 + 0.57 Re^{1/2} Sc^{1/3}\right) (1 + BM)^{-0.7}. \quad (2.31)
\]

In their model, the properties are evaluated at a reference state defined with \(A_r = \frac{1}{2}\) in Eq. 2.15, instead of \(1/3\).

Abramzon & Sirignano (1989) developed a more rigorous model, based on a boundary layer thickness analysis accounting for the boundary layer thickening induced by the Stefan flow. They define the evaporation flowrate as:

\[
\dot{m} = 2\pi \rho D_{vap} r_s Sh^* \ln(1 + BM) = 2\pi \frac{\kappa}{c_p} r_s Nu^* \ln(1 + BT). \quad (2.32)
\]

Note that the equality in Eq. 2.32 holds only if enthalpy diffusion is neglected. From Eq. 2.32 it follows that the relation between \(BT\) and \(BM\) is:

\[
BT = (1 + BM) \left(\frac{Sh^*}{Nu^*} \frac{1}{Le}\right) - 1. \quad (2.33)
\]

As previously mentioned, Abramzon & Sirignano (1989) use \(\tau_{p,vap}\) instead of \(\tau_p\) in the convective term of the gas heat equation: this carries over in Eq. 2.32, where they have \(\tau_{p,vap}\) instead of \(\tau_p\), and in Eq. 2.33, which they give as:

\[
BT = (1 + BM) \left(\frac{\tau_{p,vap}}{\tau_p} \frac{Sh^*}{Nu^*} \frac{1}{Le}\right) - 1. \quad (2.34)
\]

The modified Nusselt and Sherwood numbers are calculated as:

\[
Nu^* = 2 + (Nu_0 - 2)(1 + BT)^{-0.7} \frac{BT}{\ln(1 + BT)}; \quad (2.35)
\]

\[
Sh^* = 2 + (Sh_0 - 2)(1 + BM)^{-0.7} \frac{BM}{\ln(1 + BM)}; \quad (2.36)
\]
where any of the correlations described above can be used for $Nu_0$ and $Sh_0$. Note that the calculation is iterative, since $B_T$ also depends on $Nu^*$. The transfer rates $\dot{m}$ and $\dot{q}$ are then calculated from Eq. 2.32 and Eq. 2.14.

The Abramzon & Sirignano (1989) model is the most widely used in evaporation calculations. Ebrahimian & Habchi (2011) however show that the exclusion of enthalpy diffusion in the model leads to overprediction of the evaporation rate.

On the ground of the Abramzon & Sirignano (1989) model, a closure that accounts for enthalpy diffusion would be:

$$\dot{m} = 2\pi \rho D_{vap} r_s Sh^* \ln(1 + B_M); \quad (2.37)$$

$$\dot{q} = \frac{\dot{m}}{\nu_p} \frac{(T_s - T_\infty)}{B_T}; \quad (2.38)$$

where $B_M$ is given by Eq. 2.11, but $B_T$ is given by:

$$B_T = \int_1^{1+B_M} \phi \zeta^\phi - 1 \exp \left( \phi \left( \frac{\nu_p,vap}{\nu_p,inert} \right) (1 - Y_s) (\zeta - 1) \right) \, d\zeta; \quad (2.39a)$$

$$\phi = \frac{1}{Le} \frac{Sh^*}{Nu^*}. \quad (2.39b)$$

Evaporation in convective environment implies that circulation inside the droplet is induced by the shear on the droplet surface. A complete solution of the three-dimensional flow field inside the droplet is often impractical, therefore a number of models have been proposed to correct for liquid circulation.

In the rapid mixing model all liquid fields are treated as uniform, and Eq. 2.7 is replaced by a global energy balance, obtained by integrating Eq. 2.7 and considering that the temperature gradient is zero everywhere except at the surface:

$$\frac{r_s^2}{\alpha_l} \frac{dT_l}{dt} = \frac{\partial T_l}{\partial \omega} \bigg|_{\omega=1} = -\dot{q} + \dot{m} \Lambda \frac{1}{4\pi r_s \kappa_l}. \quad (2.40)$$

This model requires that the timescale for liquid mixing is very small, therefore it is applicable in case of very strong internal circulation or extremely small Biot number.

In the conduction limit model, internal circulation is disregarded entirely, whereby heat is transported symmetrically and purely by conduction, so that Eq. 2.7 applies without modifications. This model is likely suited to fairly big droplets, for which the surface velocity induced by the gas friction is small.

The results given by the diffusion limit model and the rapid mixing model define the two extremes bounding the possible range of real conditions.
The vortex model (Abramzon & Sirignano 1989), based on observations of droplets falling at terminal velocity, approximates the flow field inside the droplet with the Hill spherical vortex solution (Batchelor 2000):

\[ u_\omega = -U_s (1 - \omega^2) \cos \theta; \]
\[ u_\theta = U_s (1 - 2\omega^2) \sin \theta; \]

where the surface velocity \( U_s \) is estimated as:

\[ U_s = \frac{C_D}{32} |U_\infty - U_l| \left( \frac{\mu}{\mu_l} \right) Re_\infty. \]

Equation 2.7 is thus replaced by:

\[ \frac{r_s^2}{\alpha_l} \frac{\partial T_l}{\partial t} = \frac{\partial^2 T_l}{\partial \omega^2} + \frac{1}{\omega^2} \frac{\partial^2 T_l}{\partial \theta^2} + \left( \frac{2}{\omega} + \frac{1}{\kappa_l} \frac{\partial \kappa_l}{\partial \omega} + \frac{1}{2 \alpha_l} \frac{d r_s^2}{d t} - \frac{r_s u_\theta}{\alpha_l} \right) \frac{\partial T_l}{\partial \omega} \]
\[ + \frac{1}{\omega^2} \left( \frac{1}{\tan \theta} + \frac{1}{\kappa_l} \frac{\partial \kappa_l}{\partial \theta} - \frac{r_s}{\alpha_l} \omega u_\theta \right) \frac{\partial T_l}{\partial \theta}; \]

and supplied with the symmetry boundary conditions:

\[ \frac{\partial T_l}{\partial \omega}\bigg|_{\omega=0} = 0; \quad \frac{\partial T_l}{\partial \theta}\bigg|_{\omega=1} = 0; \quad \frac{\partial T_l}{\partial \theta}\bigg|_{\theta=0,\pi} = 0. \]

Equation 2.9 is replaced by:

\[ \int_0^\pi \frac{\partial T_l}{\partial \omega}\bigg|_{\omega=1} \sin \theta \, d\theta = -\frac{(q + \dot{m}\Lambda)}{2\pi r_s \kappa_l(\omega = 1, t)}. \]

In the effective conductivity model, the purely conductive formulation of Eq. 2.7 is kept, and the increased transport due to liquid circulation is accounted for with a factor \( \chi \) that enhances the conductivity \( \kappa_l \) (and thermal diffusivity \( \alpha_l \)). Abramzon & Sirignano (1989) give the following empirical correlation for \( \chi \):

\[ \chi = 1.86 + 0.86 \tanh \left[ 2.245 \log_{10} \left( \frac{P_{el}}{30} \right) \right]; \]

where \( P_{el} = \frac{2U_s r_s}{\alpha_l} \) is the droplet thermal Pécelt number, estimated on the basis of Eq. 2.42.

2.1.4. Droplet history

The droplet history can be calculated by integrating Eq. 2.8 and solving Eq. 2.7 and the gas phase simultaneously. The simplest scenario occurs for a static droplet, when its temperature is uniform and equal to the equilibrium wet bulb temperature. This condition is attained when no sensible heat is being transported in the liquid, i.e. when the heat provided by the gas balances the latent heat of evaporation (\( \dot{m}\Lambda = -\dot{q} \)). From Eq. 2.14, it follows that the wet bulb temperature can be calculated by solving the following equation:
When the droplet temperature is constant, the Spalding mass transfer number \( B_M \) and all averaged gas properties are also constant, and the droplet surface decreases linearly in time (\( \rho_2 \)-law):

\[
\frac{dr_s^2}{dt} = -K_{wb} = -\left[ 2\frac{\rho_l}{\rho_i} \overline{D}_{vap} \ln (1 + B_M) \right]. \tag{2.48}
\]

The droplet evolution is thus characterized by two regimes:

1. Initial transient heat-up or cool-down regime, until the wet bulb temperature \( T_{wb} \) is reached;
2. Evaporation at constant temperature \( T_{wb} \) (asymptotic regime: \( \rho_2 \)-law).

Here we suggest a simplified solution based on the rapid mixing assumption for the liquid phase.

2.1.4.1. Transient regime

The two cases \( T_{l0} < T_{wb} \) and \( T_{l0} > T_{wb} \) must be distinguished. If the initial droplet temperature \( T_{l0} \) is lower than the wet bulb temperature, the heat transferred from the gas to the liquid will be partly employed as sensible heat to heat the droplet up to \( T_{wb} \), and partly as latent heat of vaporization. If the initial droplet temperature \( T_{l0} \) is higher than the wet bulb temperature, the latent heat will be initially provided by the liquid and as a result the droplet will cool down to \( T_{wb} \).

If we assume the rapid mixing model for the heat transfer in the liquid and constant liquid properties we get:

\[
\tau_h \left( \frac{r_s}{r_{s0}} \right)^2 \frac{dT_l}{dt} = \left. \frac{\partial T_l}{\partial \omega} \right|_{\omega=1} = -\frac{\dot{q} + \dot{m} \Lambda}{4\pi r_s \kappa_{l0}}; \tag{2.49}
\]

where \( \tau_h = \frac{r_{s0}^2}{\alpha_{l0}} \).

**Case A: \( T_{l0} < T_{wb} \).** Substituting Eq. 2.13 and 2.14 into Eq. 2.49 and linearizing the right hand side we get:

\[
\frac{dT_l}{dt} = \frac{C_1}{\tau_h} [(T_\infty - T_l) - C_2]; \tag{2.50}
\]

whose solution is:

\[
T_l = T_\infty - C_2 - [(T_\infty - T_{l0}) - C_2] e^{-C_1 t/\tau_h}; \tag{2.51}
\]
where:

\[ C_1 = \frac{\ln(1 + B_{M0})}{Le_0} \frac{1}{B_{T0}} \frac{\kappa_0}{\kappa_{l0}}; \quad (2.52a) \]

\[ C_2 = \frac{A_0 B_{T0}}{\tau_{p0}}. \quad (2.52b) \]

The end of the transient regime is the time needed to reach the wet bulb temperature:

\[ \tau_{wb} = \frac{\tau_h}{C_1} \ln \left( \frac{(T_\infty - T_{l0}) - C_2}{(T_\infty - T_{wb}) - C_2} \right). \quad (2.53) \]

**Case B: \( T_{l0} > T_{wb} \)** We neglect the heat transferred from the gas to the droplet in Eq. 2.49 and assume that during the cool-down all the latent heat is provided by the liquid. Then, linearizing the right-hand-side, we get:

\[ \frac{dT_l}{dt} = -\frac{C_1 C_2}{\tau_h}; \quad (2.54) \]

whose solution is:

\[ T_l = T_{l0} - C_1 C_2 \frac{t}{\tau_h}. \quad (2.55) \]

In this case the end of the transient regime is at the time:

\[ \tau_{wb} = \frac{T_{l0} - T_{wb}}{C_1 C_2}. \quad (2.56) \]

Eq. 2.51 or Eq. 2.55 can be used in the vapour–liquid equilibrium relation to obtain \( Y_s(t) \) and thereby \( B_M(t) \), which can be used to calculate the droplet history by solving the simplified droplet mass balance:

\[ \frac{dr_s^2}{dt} = -2 \frac{\bar{p}_0}{\rho_{l0}} D_{vap,0} \ln(1 + B_M). \quad (2.57) \]

2.1.4.2. **Asymptotic regime (d²-law)**

After \( t = \tau_{wb} \), the droplet vaporizes at constant temperature \( T_{wb} \) according to Eq. 2.48, with the initial condition given by \( r_s^2(\tau_{wb}) \), calculated from Eq. 2.57.

The droplet history in the asymptotic regime is then:

\[ r_s^2 = r_s^2(\tau_{wb}) - K_{wb} (t - \tau_{wb}); \quad (2.58) \]

and the total droplet lifetime is:

\[ \tau_{life} = \tau_{wb} + \frac{r_s^2(\tau_{wb})}{K_{wb}}. \quad (2.59) \]

Summarizing, the droplet history is determined by the gas boundary conditions at infinity \( (T_\infty, Y_\infty) \), which determine the wet bulb temperature \( T_{wb} \).
2.1. Droplet evaporation

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and the evaporation constant $K_{wb}$, and the droplet initial conditions $(T_{l0}, r_{s0})$, which determine the time $\tau_{wb}$ to reach $T_{wb}$.

Figure 2.1 shows a comparison between the droplet history calculated with the simplified model described above, and the full solution; for a static n-heptane droplet with different initial and boundary conditions.

We note that the $d^2$-law regime is never attained for a droplet evaporating in convective environment: in this case, even at constant temperature, the evaporation rate $\dot{m}$ does not depend linearly on the droplet radius, due to the factor $Sh^*$ in Eq. 2.37, which depends on the instantaneous Reynolds number.

2.1.5. Multicomponent droplets

When the liquid components are more than one, difference in volatilities gives rise to different evaporation rates for the components, and thereby induces composition gradients and species transport in the liquid phase (Law 1982; Sazhin 2006).

An exact modelling of multicomponent species transport would require the generalized Stefan–Maxwell equations as the phenomenological relations between the diffusive fluxes $j_i$ and their driving forces in both phases (Bird et al. 2007b):

$$\tilde{x}_i \nabla \ln (\gamma_i \tilde{x}_i) = - \sum_{j=1}^{N} \tilde{x}_i \tilde{x}_j \rho \tilde{D}_{ij} \left( \frac{j_i}{x_i} - \frac{j_j}{x_j} \right); \quad (2.60)$$

where $\tilde{x}$ and $x$ are respectively the mole and mass fractions in the phase considered, $\gamma$ the activity coefficients, and $\tilde{D}$ the matrix of binary Stefan–Maxwell diffusivities.

Some attempts have been made at solving the full multicomponent formulation (Tonini & Cossali 2016), however by far the most common approach has been to approximate the multicomponent diffusion as purely Fickian.

The velocity field in the gas phase is described by Eq. 2.1. The heat transport equation reads (under the assumption of Fickian diffusion):

$$\rho c_p \frac{DT}{Dt} = \nabla \cdot (\kappa \nabla T) + \rho \sum_{j=1}^{N} c_{p,j} D_j \nabla T \cdot \nabla Y_j; \quad (2.61)$$

where $D_i$ is the diffusivity of component $i$ in the gas mixture constituted by all other components.

It is frequently assumed that the gas surrounding the droplet is insoluble in the liquid phase. The composition field in the gas phase is described by $(N - 1)$ transport equations for the mass fractions $Y_i$ of the vaporizing components (the mass fraction of the inert gas is obtained by $Y_N = 1 - \sum_{j=1}^{N-1} Y_j$):

$$\rho \frac{DY_i}{Dt} = \nabla \cdot (\rho D_i \nabla Y_i); \quad (2.62)$$
Figure 2.1: Time evolution of the normalized droplet surface area for a pure static n-heptane droplet. Comparison between full solution and simplified model.
The mass and heat fluxes integrated over the droplet surface are:

\[
\dot{m} = \int_S \left( -\rho \sum_{j=1}^{N-1} D_j \nabla Y_j + \rho \mathbf{u} \sum_{j=1}^{N-1} Y_j \right) \cdot \mathbf{n} \, dS. \tag{2.63}
\]

\[
\dot{m}_i = \varepsilon_i \dot{m} = \int_S (-\rho D_i \nabla Y_i + \rho \mathbf{u} Y_i) \cdot \mathbf{n} \, dS. \tag{2.64}
\]

\[
\dot{q} = \int_S (-\kappa \nabla T) \cdot \mathbf{n} \, dS; \tag{2.65}
\]

where \(\varepsilon_i\) is the fractional evaporation rate, and clearly \(\sum_{j=1}^{N-1} \varepsilon_j = 1\).

On the liquid side, in addition to Eq. 2.7 and Eq. 2.8, \((N - 2)\) equations for the mass fractions \(X_i\) in the liquid must be solved. In the case of a static droplet (or when a diffusion limit model is employed to account for liquid circulation in a convective environment), they read (assuming Fickian diffusion):

\[
\frac{r_s^2}{D_{i,l}} \frac{\partial X_i}{\partial t} = \frac{\partial^2 X_i}{\partial \omega^2} + \left( \frac{2}{\omega} + \frac{1}{\rho_l} \frac{\partial \rho_l}{\partial \omega} + \frac{1}{D_{i,l}} \frac{\partial D_{i,l}}{\partial \omega} + \frac{1}{2} \frac{\rho_l}{D_{i,l}} \frac{dr_s^2}{dt} \right) \frac{\partial X_i}{\partial \omega}; \tag{2.66}
\]

where \(D_{i,l}\) is the diffusivity of component \(i\) in the liquid mixture constituted by all other components.

The boundary conditions for the liquid phase at the droplet surface are:

\[
\left. \frac{\partial T_l}{\partial \omega} \right|_{\omega=1} = -\frac{\left( \dot{q} + \dot{m} \sum_{j=1}^{N-1} \varepsilon_j \lambda_j \right)}{4 \pi r_s \kappa_l (\omega = 1, t)}. \tag{2.67}
\]

\[
\left. \frac{\partial X_i}{\partial \omega} \right|_{\omega=1} = \frac{\dot{m} (X_{i,s} - \varepsilon_i)}{4 \pi r_s \rho_l (\omega = 1, t) D_{i,l} (\omega = 1, t)}; \tag{2.68}
\]

If an effective diffusivity model is employed, the liquid diffusivities are multiplied by a factor given by:

\[
\chi_i = 1.86 + 0.86 \tanh \left[ 2.245 \log_{10} \left( \frac{Pe_{i,l}}{30} \right) \right]; \tag{2.69}
\]

where \(Pe_{i,l} = \frac{2 U_s r_s}{D_{i,l}}\) is the droplet mass Pécellet number for component \(i\).

If a vortex model is employed, Eq. 2.66 take a form similar to Eq. 2.43.

If all vapour components have the same diffusion coefficient in the gas mixture, it can be shown that the same quasi–steady closures for \(\dot{m}\) and \(\dot{q}\) described for the pure droplet apply, both for static and convective conditions, provided that the Spalding transfer numbers are defined as:

\[
B_M = \frac{\sum_{j=1}^{N-1} Y_{j,s} - \sum_{j=1}^{N-1} Y_{j,\infty}}{1 - \sum_{j=1}^{N-1} Y_{j,s}}; \tag{2.70}
\]
\[ B_T = \frac{1}{Le} \int_{1}^{1 + B_M} \zeta^{(1/Le) - 1} \exp \left[ \frac{1}{Le} \left( \sum_{j=1}^{N-1} Y_{j,ref} \bar{c}_p, i - \bar{c}_p, N \right) \left( 1 - \sum_{j=1}^{N-1} Y_{j,s} \right) (\zeta - 1) \right] d\zeta. \] (2.71)

In this case the fractional evaporation rate is given by:

\[ \varepsilon_i = Y_{i,s} + \frac{1}{B_M} (Y_{i,s} - Y_{i,\infty}). \] (2.72)

Obviously the vapour components can have very different diffusivities, in which case a common approximation is (Tonini & Cossali 2015):

\[ \bar{D}_{vap} = \frac{\sum_{j=1}^{N-1} Y_{j,ref} \bar{D}_j}{\sum_{j=1}^{N-1} Y_{j,ref}}. \] (2.73)

### 2.1.6. Thermodynamic equilibrium

At the droplet surface, thermodynamic equilibrium dictates that the fugacity of each component in the liquid phase is equal to its fugacity in the vapour phase. This relation can be expressed as (Poling et al. 2000):

\[ \hat{\phi}_i(T_s, P_s, \tilde{Y}_s) \tilde{Y}_{i,s} P_s = \gamma_i(T_s, \tilde{X}_s) \tilde{X}_{i,s} P_{sat}^i(T_s); \quad i = 1, \ldots N; \] (2.74a)

\[ \sum_{i=1}^{N} \tilde{Y}_{i,s} = 1. \] (2.74b)

Here \( \hat{\phi}_i \) is the fugacity coefficient of component \( i \) in the vapour phase, \( \gamma_i \) is the activity coefficient of component \( i \) in the liquid phase, \( P_{sat}^i \) is the equilibrium vapour pressure of component \( i \) at temperature \( T_s \), \( P_s \) is the total pressure of the vapour phase, \( \tilde{Y}_s \) and \( \tilde{X}_s \) are the compositions of the vapour and liquid phase respectively, expressed as mole fractions.

The vapour pressure relation \( P_{sat}^i(T) \) is best expressed by semiempirical formulas of the type (Green & Perry 2007):

\[ P_{sat}^i(T) = P_{ref} \exp \left[ C_1 + C_2 \frac{T_{ref}}{T} + C_3 \ln \left( \frac{T}{T_{ref}} \right) + C_4 \left( \frac{T}{T_{ref}} \right)^{C_5} \right]. \] (2.75)

These are fitted expressions modelled on the basis of integration of the Clapeyron relation, which requires that (Smith et al. 2004):

\[ \frac{d \ln(P_{sat})}{d \left( \frac{1}{T} \right)} = - \frac{\tilde{\Lambda}}{R \Delta z}; \] (2.76)

where \( \tilde{\Lambda} \) is the molar latent heat of vaporization and \( \Delta z \) is the change in the compressibility factor during the phase change.
2.1. Droplet evaporation

The fugacity coefficients deviate from unity only at high pressures and/or very low temperatures, when non-ideality of the gas phase is not negligible (Ebrahimian & Habchi 2011; Mahiques et al. 2017). Their form depends on the real gas equation of state of the model. A common choice is to use a first order virial expansion as equation of state (Smith et al. 2004), in which case the fugacity coefficients are given by:

\[
\ln \hat{\phi}_i = \left(2 \sum_{j=1}^{N} \tilde{Y}_j B_{ij} - B\right) \frac{P}{RT}; \quad (2.77a)
\]

\[
B = \sum_{i=1}^{N} \sum_{j=1}^{N} \tilde{Y}_i \tilde{Y}_j B_{ij}. \quad (2.77b)
\]

The binary second virial coefficients \( B_{ij} \) depend on temperature only and can be predicted by correlations such as those by Hayden & O’Connell (1975) or Tsonopoulos (1974).

The activity coefficients account for the molecular interactions of the components in the liquid phase (thus for a pure liquid \( \gamma = 1 \)). Their importance varies widely depending on the liquid mixture. For instance, many mixtures of linear hydrocarbons of similar molecular weight, exhibit \( \gamma_i \approx 1 \) over the whole range of compositions, as the molecules are sufficiently similar. As a consequence, their vapour–liquid equilibrium is well predicted by Raoult’s law. On the other hand, mixtures of highly different molecules, like water and an organic compound, tend to have strongly asymmetric interactions, and display non-ideal equilibrium features like azotropic points that can be predicted only by an accurate model for \( \gamma_i \) (see Fig. 2.2 for two examples of both tendencies).

There are several semiempirical correlations for the prediction of activity coefficients as functions of temperature and composition. Prausnitz et al. (1999) discuss the merits and applicability of each model at length.

The UNIQUAC model gives good predictions for a wide range of mixtures; it is based on pure component parameters that represent differences in size and shape of the molecules in the mixture, and binary parameters that represent energy interactions of pairs of components (Poling et al. 2000):

\[
\ln \gamma_i = \ln \left(\frac{\Phi_i}{X_i}\right) + \frac{z}{2} q_i \ln \left(\frac{\theta_i}{\Phi_i}\right) + l_i - \frac{\Phi_i}{X_i} \sum_j \tilde{X}_j l_j 
\]

\[
+ q_i \left[1 - \ln \left(\sum_j \theta_j \tau_{ji}\right) - \sum_j \frac{\theta_j \tau_{ij}}{\sum_k \theta_k \tau_{kj}}\right]; \quad (2.78a)
\]

\[
z = 10; \quad (2.78b)
\]

\[
l_i = \frac{z}{2} (r_i - q_i) - (r_i - 1); \quad (2.78c)
\]

\[
\theta_i = \frac{q_i}{\sum_j q_j \tilde{X}_j}; \quad (2.78d)
\]
\[ \Phi_i = \frac{r_i}{\sum_j r_j X_j}; \quad (2.78e) \]

\[ \tau_{ji} = \exp \left( -\frac{u_{ji} - u_{ii}}{RT} \right). \quad (2.78f) \]

The pure component parameters \( r_i \) and \( q_i \) are, respectively, measures of molecular van der Waals volumes and molecular surface areas. For each pair of components, there are two binary interaction parameters \( \tau_{ij} \) and \( \tau_{ji} \) (they are not symmetric), which are best evaluated by fitting the molecular interaction energies \( u_{ij}, u_{ii} \) and \( u_{jj} \) to experimental data as functions of temperature.
2.1. Droplet evaporation

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**Figure 2.2**: Isobaric vapour–liquid equilibrium phase diagrams for the systems n-heptane–n-decane and ethanol–water.
2.2. Droplet motion

The equation of motion for a small sphere whose mass is changing in time due to phase change, moving in a surrounding fluid of velocity $u_\infty$, reads:

$$\frac{du_t}{dt} = \frac{3 C_D \rho_\infty}{8 r_s \rho_l} (u_\infty - u_t) |u_\infty - u_t| + \left( 1 - \frac{\rho_\infty}{\rho_l} \right) g; \quad (2.79)$$

where the effects of added mass and of the Basset history term (Maxey & Riley 1983) have been neglected. The drag coefficient $C_D$ lumps together the effects of viscous drag and pressure drag. The net thrust generated by the mass expelled by the sphere is zero because mass is expelled from the surface uniformly in all directions (Crowe et al. 2012).

A variety of correlations for the drag coefficient have been used in the literature for evaporating droplets. Renksizbulut & Haywood (1988) suggest:

$$C_D = \left( \frac{24}{Re_d} + 4.8 Re_d^{-0.37} \right) (1 + B_T)^{-0.2}; \quad (2.80)$$

valid for droplet Reynolds numbers in the range $10 < Re_d < 300$, which is the relevant range in most applications. The droplet Reynolds number is defined with the free stream gas density, but the viscosity of the evaporating film, which is thus calculated at the reference conditions given by Eq. 2.15:

$$Re_d = \frac{2r_s |u_\infty - u_t| \rho_\infty}{\mu}. \quad (2.81)$$

With its dependence on the heat transfer number $B_T$, Eq. 2.80 is one the few available correlations that account for the reduction of friction drag and increase of pressure drag cause by surface evaporation, an effect akin to surface blowing.
Chapter 3

Interface resolved numerical simulations with phase change

In the previous chapter, we presented an overview of droplet evaporation models that are used in numerical simulation of spray flows, when the dispersed phase is represented in a coarse–grained fashion by a collection of material points. This technique is obviously binding for droplets that are smaller than the cell size of the grid by which the computational domain is discretized, a condition that may be violated for simulations with a high desired accuracy (e.g. LES or DNS calculations).

Numerical methods that are able to resolve the gas–liquid interface do not need modelling for the exchange of mass, momentum and energy between the continuous and the dispersed phase: as a consequence they can be used in a broader range of conditions, at the expense of higher computational cost.

These methods can be sorted into two main classes: interface tracking methods and interface capturing methods. Interface tracking methods describe the interface directly as part of the computational mesh, either in a eulerian–eulerian framework, where a moving mesh that conforms to the interface is employed, or in a eulerian–lagrangian framework, where a separate lagrangian mesh (marker points) is used to track the interface on top of the underlying eulerian mesh. An advantage of these methods is the accuracy of the interface shape calculation, which is desirable in problems with phase change, where heat and mass flux vectors at the interface drive the physics; while a disadvantage is the difficulty in treating cases of large interface deformation.

Interface capturing methods describe the fluid phases by an artificial scalar field (marker function) for which a transport equation is solved. The position of the interface is inferred by the value of the marker function. These methods are well suited for the treatment of phenomena where the interface undergoes severe deformation and topological change (e.g. phase breakup and coalescence), since they contain no assumption about the connectivity of the interface, but as a disadvantage the interface shape needs to be artificially reconstructed, leading to loss of detail for quantities such as curvature (a drawback that particularly affects volume of fluid methods), which can undermine mass conservation between the fluid phases especially when phase change is present (a known issue of level set methods).
A key requirement of any interface resolved numerical method that aims at solving a multiphase flow with phase change, is that it must be able to correctly handle the discontinuities that arise due to the mass transport across the interface, i.e. the phase change. When mass is transported across the interface, the latter is not a contact discontinuity, where normal velocity would be continuous, but rather a “shock”, for which conservation of total mass, momentum and energy leads, in the incompressible limit, to the following jump conditions, also called Rankine–Hugoniot conditions (Joseph & Renardy 1993; Matalon & Matkowsky 1982):

\[
\begin{align*}
\mathcal{J} \rho (\mathbf{u} \cdot \mathbf{n} - w) &= 0; \\
\mathcal{J} \rho \mathbf{u} (\mathbf{u} \cdot \mathbf{n} - w) + p \mathbf{n} - \mu \left( \nabla \mathbf{u} + \nabla \mathbf{u}^T \right) \cdot \mathbf{n} &= -\kappa \sigma \mathbf{n} - (\nabla \sigma \times \mathbf{n}) \times \mathbf{n}; \\
\mathcal{J} [T] &= 0;
\end{align*}
\] (3.1) (3.2) (3.3)

where \( w \) is the velocity of the interface, \( \sigma \) the surface tension, \( \kappa \) the signed interface curvature, and \([g] = (g^+ - g^-)\) denotes the jump of the quantity \( g \) across the interface. Jumps across the interface can be numerically realized in essentially two ways. The jump for a field can be interpreted as a continuous, three-dimensional forcing acting across the interface on the transport equation for said field, rather than a boundary value condition on the interface, For instance, surface tension can be locally added as a body force to the Navier–Stokes equation, in a layer of prescribed thickness across the interface, and it will thus generate a stress jump across the layer. In the same fashion, a velocity divergence source can be added to the continuity equation, and it will generate a velocity jump across the layer. This concept is known as Continuous Surface Forcing (CSF) (Brackbill et al. 1992), and its main drawback is that the realization of jump conditions is not sharp, but smeared across the volume where the forcing is applied. The second approach, known as Ghost Fluid (Fedkiw et al. 1999), realizes the jump as an actual boundary value condition on the interface, by appropriately extending the field for each phase on ghost cells located on the other side of the interface (i.e. where the other phase is physically located).

In the following, we provide a brief description of the methods that are prevalent in the simulation of multiphase flows with phase transition: the Volume of Fluid (VOF) method and the Level Set method are interface capturing methods; the Front Tracking method is an interface tracking method. In recent years, the Lattice–Boltzmann method has also been used to tackle flows with phase change (Ledesma-Aguilar et al. 2014; Albernaz et al. 2015). The mathematical framework of this method is not based on transport equations discretized on a computational grid, as all the methods listed above, but on mesoscopic kinetic equations for fluid pseudoparticles embedded in a regular lattice; therefore its description is out of the scope of this work.

A short description of the Immersed Boundary method closes the chapter. This interface tracking method, which has been used extensively to simulate
3.2. Level set

Volume of Fluid (VOF) methods treat the multiphase flow in a one-field, fully eulerian formulation, using marker functions to represent the fluid phases (Prosperetti & Tryggvason 2007).

A multiphase problem with $N$ phases is solved on a eulerian grid with the aid of transport equations for $(N - 1)$ volume fractions $f_j$, which represent the amount of each phase in each computational cell. Therefore $0 \leq f_j \leq 1$. Physical and transport properties are weighted in the appropriate way with $f_j$, or the corresponding mass/mole fractions. A cell where no volume fraction $f_j$ has a value of 1 contains one or more interfaces.

Solution for $f_j$ provides the position of each interface, but not its shape. Therefore the geometric details (typically orientation and curvature) must be reconstructed in each cell using the neighbouring values of $f_j$. Reconstruction techniques for the interface are reviewed in Scardovelli & Zaleski (1999) and Prosperetti & Tryggvason (2007).

Welch & Wilson (2000) developed a VOF method for flows with phase change driven by thermal gradients, e.g. boiling flows.

Schlottke & Weigand (2008) developed a VOF method for the simulation of evaporating and deforming droplets, where phase change is driven by species concentration gradients, whereby they employed two volume fractions, $f_1$ and $f_2$, for the liquid and vapour phase respectively, and used $f_1$ to reconstruct the interface separating the liquid from the gas/vapour mixture.

Banerjee (2013) simulated the evaporation of a single multicomponent droplet with the VOF method.

3.2. Level set

In Level Set methods, the interface is defined as the zero level curve of a level set function $\phi$, which represents the signed distance form the interface. This function is advected by its velocity $\mathbf{w}$, which is the sum of the fluid velocity and the interface displacement speed caused by the phase change.

In order to reconstruct the geometrical features of the interface from the values of $\phi$, it is crucial that the level set function stays indeed a signed distance function as time integration advances, at least in the vicinity of the interface. This condition is not guaranteed automatically by the transport equation for $\phi$ (Prosperetti & Tryggvason 2007); therefore, every few time steps, the level set function $\phi$ needs to be reinitialized as a new function $\phi_d$ that keeps the same zero level curve of $\phi$, but satisfies the constraint $|\nabla \phi_d| = 1$ over an appropriately selected vicinity of the interface. This constraint guarantees that $\phi_d$ is a distance
function, because it implies that $\nabla \phi_d \cdot \mathbf{n} = 1$, where $\mathbf{n}$ is the unit vector normal to the interface.

While the interface shape details can in principle be reconstructed with any given level of accuracy just by extending their computational stencil around the zero level curve (provided that the stencil is defined in a region where $\phi$ is a signed distance function), it is well documented that level set methods can generate mass loss in under-resolved regions.

Nguyen et al. (2001) developed a general Level Set method for treating two-phase incompressible flow where the interface is “reactive”, i.e. one phase is being converted into the other, as in premixed combustion or phase transition. The robustness of their method relies on coupling the level set formulation with the Ghost Fluid method of Fedkiw et al. (1999), which they use to avoid smearing of the jump conditions around the interface and achieve a sharp interface representation.

Wang et al. (2004) used a Level Set coupled with Ghost Fluid method to simulate two-dimensional heterogeneous solid-gas propellant combustion.

Gibou & Fedkiw (2005) applied the same concept to the simulation of Stefan flow caused by phase change driven by a temperature gradient.

Tanguy et al. (2007) extended the method to the treatment of vaporizing two-phase flow with high density ratio between the two phases, by improving the liquid velocity field extension in the ghost cells.

### 3.3. Front tracking

In front tracking methods the interface is represented by a set of connected marker points, that form a two-dimensional front contained in the underlying eulerian grid. The front is advected in a lagrangian fashion (Prosperetti & Tryggvason 2007).

The interface geometry is completely defined by the marker points set, unlike interface capturing methods where the geometric features have to be reconstructed from the values of a marker function in the eulerian cells that are close to the interface. Conversely, in front tracking methods a phase indicator function, that tells which phase is present in each eulerian cell, must be reconstructed from the front position and geometry, while in interface capturing methods this information is already carried by the marker function.

Topological change does not happen automatically, as in interface capturing methods, but has to be artificially enforced if needed when certain conditions are satisfied (e.g. merging of two fronts, or two parts of the same front, can be imposed when the two come within a specified distance).

Since the front inhabits a separate grid, at each time step information needs to be passed from the moving lagrangian front to the fixed eulerian grid (smoothing), and vice versa (interpolation). This is commonly achieved with the Immersed Boundary technique, by which any boundary condition or force that acts on the interface is smoothed to the eulerian grid by a distribution
function that has a finite support in the vicinity of the front. Although it is not necessary to use the same distribution function, this is commonly also utilized to interpolate the field values that are needed for the front advection onto the lagrangian front from the fixed eulerian grid. Note that the smoothing part of this technique is effectively a continuous surface forcing method for the eulerian grid.

Juric & Tryggvason (1998) were the first to apply a front tracking method to phase change, by simulating two-dimensional film boiling, where the phase change is driven by a temperature gradient.


3.4. **Immersed boundary method for evaporating spherical droplets**

Immersed Boundary methods represent phase boundaries with a set of lagrangian marker points, while the flow equations are solved on a fixed eulerian grid. Extra forces, calculated on the marker points, are added to the right hand side of the flow equations in order to mimic the boundary conditions at the interface. These forces are distributed to the eulerian cells that are close to the marker points by an appropriate distribution function. This concept is quite general and exploited by several numerical methods. For instance, front tracking methods rely on an immersed boundary representation of the interface, as explained in the previous section.

Immersed boundary methods can be generally classified in two categories based on the implementation of the boundary forces: in continuous forcing methods the forces are added to the governing equations prior to discretization; in discrete (or direct) forcing methods the forces are introduced after the governing equations are discretized, and are thus dependent on the numerical scheme.

In recent years immersed boundary schemes have enjoyed popularity for the simulation of particle-laden laminar and turbulent flows: the use of a uniform cartesian mesh as the eulerian grid allows for efficient and easily parallelizable computational algorithms, while for solid non-reacting and non-deforming particles the surface marker points move rigidly with the center of mass, so that no front tracking is needed.

The method originally developed by Uhlmann (2005) for particle-laden flows has been improved by Breugem (2012), making it second order accurate in space. A number of studies have been carried out with this method, for different particle sizes (Costa et al. 2016), particle shapes (Ardekani et al. 2016), Reynolds numbers (Lashgari et al. 2014), solid volume fractions (Picano et al. 2014).
2015), and solid–liquid density ratios (Fornari et al. 2016), showing its reliability in capturing the four–way coupling physics of the two–phase flow.

Ardekani et al. (2017) further developed the method by allowing heat transfer.

The affinity of particle–laden flows with evaporating sprays made of small spherical droplets is evident. It is then the purpose of Paper 2 in this thesis to implement in the Immersed Boundary method described in this section the coupled heat, species and momentum transfer necessary to describe phase change. Some preliminary simulations with heat transfer and without phase change have been performed; Fig. 3.1 shows an example of the temperature, velocity and droplet distribution fields that can be obtained.
Figure 3.1: Snapshots of turbulent channel flow with cold ($T = 0$) non—evaporating droplets and hot ($T = 1$) top and bottom walls. $Re_{bulk} = 5600$; density ratio = 32, liquid volume fraction = 5%, total number of particles = 84530. Temperature contours and the droplets lying on a vertical plane along the streamwise direction are shown in the top picture. The bottom picture is a zoom-in on a vertical plane perpendicular to the streamwise direction and close to the bottom wall: velocity vectors are shown on top of temperature contours. The droplets are coloured by their temperature.
Chapter 4

Conclusions and outlook

The present work is a contribution to the rapidly expanding field of numerical simulation of multiphase flow with phase change. We focused on the evaporation of small droplets, such as those occurring in spray drying, aerosols, spray combustion, and most recently flame spray pyrolysis.

We propose an adjustment of the classic vaporization expressions by Abramzon & Sirignano (1989) to account for enthalpy diffusion in the gas phase, for both convective and non-convective gas environment. Validation in the convective environment case is needed and will be performed in the future.

We suggest a simplified model for the evaporation history of a single pure droplet in non-convective environment, based on an estimate of the duration of the initial transient heat-up/cool-down.

In paper 1 we performed detailed calculation of the evaporation of a single pure and two-component droplet with variable liquid and gas properties at atmospheric pressure in non-convective environment:

- We showed that the contribution of enthalpy diffusion in the gas phase is decisive in predicting the correct evaporation rate.
- We investigated the droplet evaporation regimes for a non-ideal liquid mixture (ethanol–water), identifying, in the case of azeotropic vapour–liquid equilibrium, a range of liquid compositions where the evaporation dynamics show a pure droplet behaviour (azeotropic evaporation regime).
- For a pure droplet, the evaporation rate in the $d^2$-law regime is determined by the equilibrium wet bulb temperature, which depends on the gas boundary conditions at infinity, but not on the liquid initial conditions. We showed that on the other hand, for a two-component droplet, the value of the pseudo-equilibrium temperature during the azeotropic evaporation regime also depends on the liquid initial conditions.
- We showed that the gas phase surface concentration of each vaporizing component and its fractional evaporation rate follow different dynamics. While the former is directly related to the total vapour pressure on the surface and thus to the droplet surface temperature history, the latter is related to the liquid surface concentration, and thus evolves along
with the balance between the relative volatility of the component and its diffusional resistance in the liquid transport.

- We explored the feasibility of a simplified model that neglects the difference in volatility of the liquid components, as well as the variation in liquid properties.

Targets of future work will be the influence of higher ambient pressure, different liquid mixtures with possible extension to the treatment of emulsions, the effect of a multicomponent formulation of the mass diffusion fluxes, and the impact of the vaporization model on laminar spray combustion calculations.

In paper 2 we extend the Immersed Boundary method of Breugem (2012), originally developed for direct numerical simulation of particle−laden flow, to allow the treatment of heat and mass exchange between the continuous phase and the dispersed phase. In this way we obtain an efficient framework for interface resolved simulations of evaporating sprays made of small spherical pure component droplets. We model the heat transfer inside the droplets with an approximate analytic solution, although extension to a fully resolved liquid phase is straightforward if needed. No modelling is needed for the heat and mass transfer rates, since the calculations are interface resolved.

Future work will be devoted to further validation of the method and study of droplet ensemble evaporation in laminar and turbulent channel and duct flow.
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