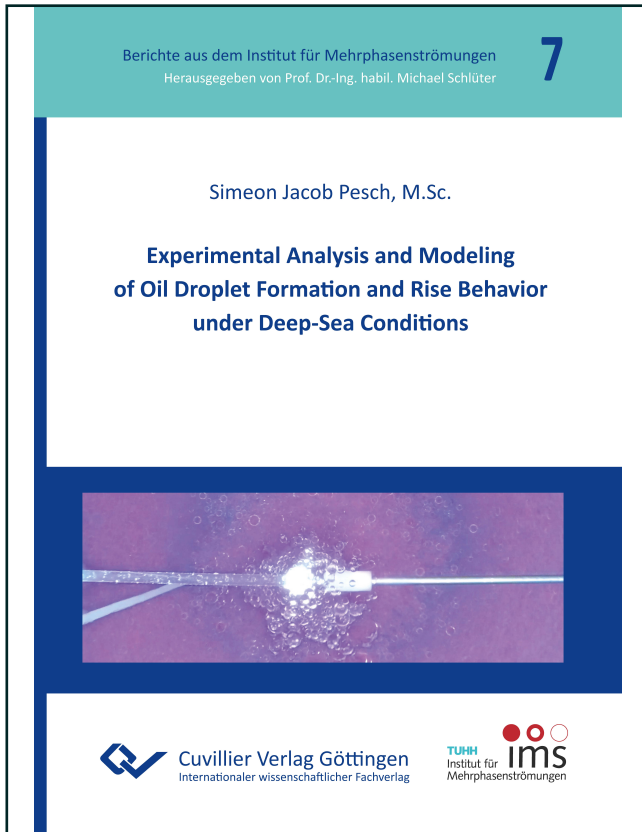




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**Experimental Analysis and Modeling of Oil Droplet  
Formation and Rise Behavior under Deep-Sea  
Conditions**



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# Chapter 1

## Introduction

Multiphase processes play a critical role in many industrial applications. The turbulent dispersion and mixing in, for instance, chemical reactors enable enhanced heat and mass transfer and thus contribute crucially to a safe and economic process operation. The rise or sedimentation behavior of (fluid) particles decides upon the flow structures and residence times in, for example, absorption columns and hence significantly influences the species transfer. While many of these processes and phenomena have been investigated and mathematically described for decades, there is still a considerable need for further research, particularly when complex substance systems or extreme process conditions come into play.

The importance and need for research with regard to multiphase flow is however not limited to industrial processes. The explosion of the oil exploration rig *Deepwater Horizon* (DWH) in the Gulf of Mexico in April 2010, which marked the beginning of one of the most devastating marine environmental disasters in the history of mankind, was followed by a subsea oil and gas blowout of unprecedented scale. The multiphase oil and gas free jet emanating from the broken wellhead, supplemented by the never before tested subsea application of chemical dispersants, could not be stopped for 87 days. The extreme environmental conditions (high pressure, low temperature) in the deep sea raised extreme challenges for both the spill response and the modeling of the dispersion caused by the blowout as well as the subsequent rise and distribution behavior of the droplets and bubbles in the water column.

While the existing model correlations from chemical engineering can provide a good basis for the fluid mechanical modeling of deep-sea oil spills, some particularities of the substance system and the ambient conditions in the deep ocean require special attention: The large amount of natural gas, partly dissolved in the crude oil, may affect the dynamics of both the oil dispersion and the drop rise, as the solubility is a function of pressure. Moreover, the subsea application of chemical dispersants further complicates the already complex substance system. Also, the very complex multiphase flow structure depends on the irregular shape

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of the damaged blowout preventer (BOP), a safety device that failed during the *Deepwater Horizon* incident and caused an additional pressure drop at the blowout site. And finally, as is often the case in engineering too, the scale-up from lab scale to field scale requires careful validation.

To account for these special features and uncertainties of deep-sea oil spills, in the scope of this thesis, several specially-designed experimental facilities and measurement protocols are developed and employed. The experimental investigations encompass the measurement of substance properties, the analysis of the dispersion behavior and resulting droplet size distributions in free jets, and investigations of phase behavior and drop rise in counter-current flow. Experimental conditions range from ambient to artificial deep-sea conditions and both a surrogate material system, which is harmless to health and the environment, and gas-saturated crude oil and seawater are used. The size of the experimental facilities ranges from lab scale to pilot-plant scale in order to enable a proper validation of the scale-up in multiphase free jets. Model correlations for the prediction of droplet sizes and the rise behavior are developed and validated. An existing far-field oil distribution model is improved by means of a new module, enabling an instantaneous 3-D oil spill simulation considering the effects, which are experimentally discovered and modeled in the scope of this work.

This thesis presents work that is a part of the research project "Center for the Integrated Modeling and Analysis of Gulf Ecosystems" (C-IMAGE), financed by the "Gulf of Mexico Research Initiative" (GoMRI). It aims to answer three major open questions that are to date very controversial in the oil spill research community:

1. Which droplet sizes occur during a deep-sea blowout?
2. How fast are the droplets rising? (And where does the oil end up?)
3. Was "Sub-Surface Dispersant Injection" (SSDI) necessary in the case of the *Deepwater Horizon* spill? (Or would it be in different oil spill scenarios?)

# Chapter 2

## State of Knowledge

This chapter provides the theoretical background that is required to understand the developments and findings of this work. First, the state of knowledge with respect to subsea crude oil production and accidental oil releases as well as the modeling approaches for blowout and oil fate simulations available in literature are briefly explained. Then, the physical processes involved in multiphase free jets, droplet breakup and the buoyant motion of fluid particles are detailed.

### 2.1 Subsea Oil Production and Oil Spills

Despite the increasing development of renewable energies, the global demand for fossile energy resources is consistently high. To date, crude oil is the most important energy source, followed by coal and natural gas [Int19]. However, most easily accessible onshore oil production sites are depleted. Hence, the exploitation of more remote deposits, for instance drilling operations in Arctic regions and offshore production in deep-sea areas, become profitable. The production from remote oil reservoirs, especially beneath water-column depths of multiple hundred or even several thousand meters, creates new technical challenges and additional risks due to the poor accessibility of the production sites and the extreme local environmental conditions.

The possible consequences became evident in April 2010, when the oil exploration rig *Deepwater Horizon* (DWH) exploded and sank in the north-eastern Gulf of Mexico. The implemented safety measures failed, leading to a major deep-sea oil well blowout of approximately 750 million liters of crude oil and huge amounts of natural gas being spilled into the ocean over a period of 87 days at a water depth of 1.524 meters [McN12]. The DWH spill was unprecedented by both the volume of hydrocarbon release and the depth of discharge. An estimated 11,000 km<sup>2</sup> of ocean surface and 2,000 km of coastline were

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impacted [Mac15, Kuj20]. Earlier major spills like the *Exxon Valdez* accident near the coast of Alaska in 1989 (oil tanker crash, surface spill) and the *Ixtoc I* spill in the southern Gulf of Mexico in 1979 (oil well blowout, water depth: 50 m) occurred in much shallower regions than the DWH spill. The latter is a first-of-its-kind example of an ultra-deep oil spill. The DWH spill is particularly relevant with regard to recent drilling operations penetrating ever deeper waters [Mur20a].

### 2.1.1 Oil Reservoirs

The conditions inside an oil reservoir determine the crude oil properties and composition as well as the circumstances in terms of temperature, pressure, pressure gradients, etc., under which the oil production or an oil well blowout occur. Reservoir systems are geological settings that are able to develop and store hydrocarbons. A crude oil system has to fulfill several physical and chemical conditions in order to form a reservoir. The rock or sediment must contain some percent of organic material and exhibit a certain porosity in order to enable migration of the built hydrocarbons. On the other hand, an impermeable layer is required in order to trap the relatively light oil and gas and thus prevent it from ascending. A high temperature is important for the chemical maturation of the fossil organic material by thermal cracking into liquid and gaseous hydrocarbons, which typically takes place over several ten million years. The pressure in oil reservoirs is typically very high due to the hundreds to thousands of meters thick layers of rock (and possibly water) and the formation of hydrocarbons [Huc04, Sat16].

In the porous rock of an oil reservoir, different fluid phases can be present. In *undersaturated oil reservoirs*, no free gas is present under the respective (initial) reservoir pressure while in *saturated oil reservoirs* the reservoir pressure is equal to the bubble-point pressure, leading to a full gas-saturation of the liquid oil [Ahm10]. If the reservoir pressure is below the bubble-point pressure of the system, the reservoir is referred to as a *gas-cap reservoir*, as is the case for the *Macondo* reservoir, where the DWH spill occurred [Hic12]. Here, three phases are present: crude oil, natural gas, and formation water. Due to the different densities of these phases, they are stratified in three layers, but the phases are not necessarily sharply separated. Water is the heaviest component and hence forms the bottom layer, while the top layer is made up of gas. The oil is located between these two layers [Sat16].

### 2.1.2 Types and Properties of Crude Oil

Crude oils are complex organic mixtures of various components. Their physical and chemical properties depend on the specific molecular composition of the hydrocarbons, which in turn

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depend on the source biomass type and age, as well as the reservoir location and conditions, as explained above. A typical crude oil composition comprises saturates, aromatic hydrocarbons, resins, asphaltenes, and dissolved gaseous components of varying carbon numbers. A crude oil with a high sulfur content is referred to as *sour* crude oil, as opposed to *sweet* crude oil, which has a fairly low sulfur content [Old20, Tre17].

As most of the substance properties of crude oil are dependent on the temperature and pressure conditions in question, they are referred to as pressure-volume-temperature (PVT) properties. The most relevant physical properties of crude oil with respect to oil production and the investigation of oil spills are the viscosity, the density (and potentially swelling due to gas saturation), the compressibility, the bubble point pressure (and gas solubility), and the interfacial tension (when brought into contact with, for instance, water) [Old20, Sat16]. Except for the density, standard SI or imperial units are used for the substance properties. The density is most often indicated as API gravity (*American Petroleum Institute*):

$$API = \frac{141.5}{sg} - 131.5, \quad (2.1)$$

with  $sg$  the specific gravity at stock tank conditions (density  $\rho$  at atmospheric pressure and 60 °F  $\equiv$  288.7 K). The API gravity is used for the classification of crude oils: Oil with an API gravity above 31.1 is considered a *light crude*, while oil with an API gravity below 22.3 is considered a *heavy crude*. Since this representation of the density is unphysical and does not allow for a statement on the density at varying p,T conditions, the usual density  $\rho$  is used in the scope of this thesis [Old20, Pes20c, Sat16].

Due to the local conditions in oil reservoirs, production pipelines or, in case of a blowout, in the deep sea, the hydrocarbon mixtures may behave significantly differently than at atmospheric conditions. The specific temperature conditions and particularly elevated pressures lead to huge variations in the oil composition and phase behavior, since the gas-in-oil solubility rises drastically with increasing pressure (and decreasing temperature) [Jag17, Old20, Pes18]. Also, the physical properties of the oil, namely density and viscosity as well as its interfacial tension against water, depend on the respective oil composition, see also chapter 4.2 and appendix C. For this reason, the p,T conditions and the related concentration of dissolved hydrocarbon gases must be accounted for, when oil reservoir characteristics, novel production techniques or submarine oil spills are investigated experimentally or numerically. The gas-saturated crude oil at the local, high-pressure conditions is referred to as “live oil” in contrast to crude oil at atmospheric conditions containing almost no C<sub>1</sub>–C<sub>4</sub> hydrocarbons, which is called “dead oil”. The design of realistic laboratory experiments under high-pressure conditions therefore includes pre-saturation of the liquid oil with natural gas or components of it in order to recombine “live oil” artificially [Pes20b, Pes20c].

### 2.1.3 Gas Solubility and Phase Behavior of Live Oil

The solubility or saturation concentration of gaseous components in crude oil depends on the temperature and particularly on the (partial) pressure. A simplified linear dependency according to Henry's law

$$x_i = H_i \cdot p_i \quad (2.2)$$

accounts for the pressure dependency, with the dissolved molar fraction  $x_i$  of the component  $i$  and its partial pressure  $p_i$  inside a virtual or actual gas phase that is in contact with the oil. The Henry constant  $H_i$  is a measure of the gas-in-oil solubility for the specific substance system and can be determined experimentally [Pfe04, Sat12]. By adjusting the numerical value and unit of  $H_i$ , the same linear relationship can also be applied to the concentration  $c_i$  instead of the molar fraction. As Henry's law is only exact for low amounts of the dissolved component corresponding to low (partial) pressures, deviations from the linear dependency can occur at high-pressure conditions [Zhe03]. The concentration of the dissolved component can therefore be supplemented by the activity coefficient  $\gamma_i$  as follows:

$$x_i \cdot \gamma_i = H_i \cdot p_i. \quad (2.3)$$

The activity coefficient results from intermolecular interactions within the liquid phase and can be described by means of  $g^E$  models, which yield the excess enthalpy  $g^E$ . The latter is defined as

$$g^E = R \cdot T \cdot \sum_i (x_i \cdot \ln \gamma_i), \quad (2.4)$$

with the universal gas constant  $R$  and the temperature  $T$  [Pfe04, Sat12]. For the small, non-polar molecules of methane that are dissolved in crude oil, the deviation from Henry's law is small even at relatively high pressures and the fit of the modified Henry's law including the activity coefficient to experimental data appears not beneficial as compared to the standard Henry's law, also with regard to other, larger error sources [Old20, Pes18].

Depending on the specific properties of the hydrocarbon mixture, each reservoir features a unique phase diagram. An exemplary phase diagram is displayed in figure 2.1, where the reservoir pressure and temperature are plotted on the axes. The phase envelope separates the two-phase region from the single-phase liquid and vapor/gas regions, respectively. The phase envelope represents the miscibility gap with varying phase fractions as denoted by the isolines depicting the liquid fraction. The top left edge of the phase envelope corresponding to 100 % liquid oil is called the *bubble point curve*. Here, full gas saturation of the live

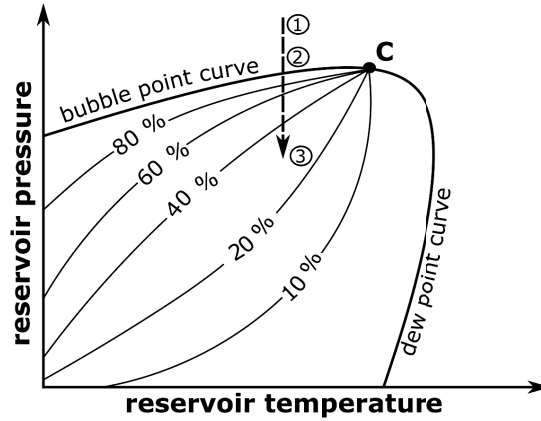


Fig. 2.1 Typical phase diagram of an oil reservoir showing the phase envelope. **C**: critical point. Isolines correspond to the liquid phase fraction. Position 1: undersaturated oil. Position 2: gas-saturated oil. Position 3: two coexisting phases or supersaturated oil; according to [Sat16].

oil is present. The right edge of the two-phase region is referred to as the *dew point curve*, corresponding to (just) pure vapor/gas. Both curves coincide at the critical point, which is marked by the **C** in figure 2.1. At and above this point, no distinguishable phases exist. Above the bubble point curve, oil is undersaturated, see position (1) in figure 2.1. If the pressure is (isothermally) reduced until the bubble point curve is reached, the oil is gas-saturated, corresponding to position (2) in the same figure. Further pressure reduction leads to the occurrence of a gas phase that consists largely of light hydrocarbons. Then, a liquid and a gaseous phase coexist in the two-phase region, see position (3) in figure 2.1 [Sat16].

Starting from an (undersaturated) single-phase liquid state (1), the pressure may reach values below the bubble point curve (3) without immediately forming a second, gaseous phase. The reason for this supersaturation (corresponding to the pressure difference between the positions (2) and (3)) is an energy barrier that has to be overcome during the so-called *nucleation*, that is the formation or re-activation of bubble nuclei within a supersaturated, homogeneous liquid [Bla75, Bau02]. The Laplace pressure (difference)

$$\Delta p = \frac{2 \cdot \sigma}{r_{\text{crit}}} \quad (2.5)$$

results from the interfacial tension  $\sigma$  and prevents growth of bubble nuclei below a critical radius  $r_{\text{crit}}$  [Mal18b]. Different approaches are available in literature, which predict the



pressure difference or critical supersaturation that is required for bubbles to form and grow [Bau02, Bla75, Old20, Pes20b, Tre17].

### 2.1.4 Oil Spill Modeling

In the following, approaches and correlations for the modeling of deep submarine oil well blowouts as well as simulation methods for the numerical investigation of the oil distribution in the aftermath of such events are briefly discussed. An overview of relevant phenomena is provided in the conceptual model sketch in figure 2.2 with special emphasis on the deep-sea conditions (high pressure, low temperature). Crucial subjects of oil spill models are the oil dispersion and the resulting droplet sizes. While large oil droplets and gas bubbles are rapidly rising upwards, smaller droplets can remain subsea and be transported laterally in so-called intrusion layers. This is accounted for by near-field models, that can also take hydrate formation and oil sedimentation into consideration. Far-field models are used to simulate the oil transport throughout the ocean and can, among other things, account for pressure-dependent partitioning and biodegradation of hydrocarbons. Subsequently, often used dimensionless numbers are introduced. Then, modeling approaches for the four stages of oil spill modeling are briefly introduced: oil dispersion during a blowout, near-field oil propagation, oil rise behavior, and oil distribution in the far field.

#### Dimensionless Numbers

To date, several approaches for the prediction of the droplet size distribution or characteristic droplet sizes thereof are available in literature. Many of these approaches are based on dimensionless numbers [Bra13, Cal86a, Cal86b, Li17, Joh13, Soc15, Wan86], particularly the Weber number

$$We = \frac{\rho_c \cdot U_0^2 \cdot D_0}{\sigma} \quad (2.6)$$

with the jet exit velocity  $U_0$ , the pipe diameter  $D_0$ , the density of the continuous phase  $\rho_c$ , and the interfacial tension  $\sigma$ . Often, in order to account for the possible influence of the dispersed phase viscosity, a second dimensionless number is used. This number can be, for example, the Reynolds number

$$Re = \frac{U_0 \cdot D_0 \cdot \rho_d}{\eta_d}, \quad (2.7)$$

with the dispersed phase density  $\rho_d$  and viscosity  $\eta_d$ , when defined for the outflow conditions of the dispersed phase. Alternatively, the viscosity number

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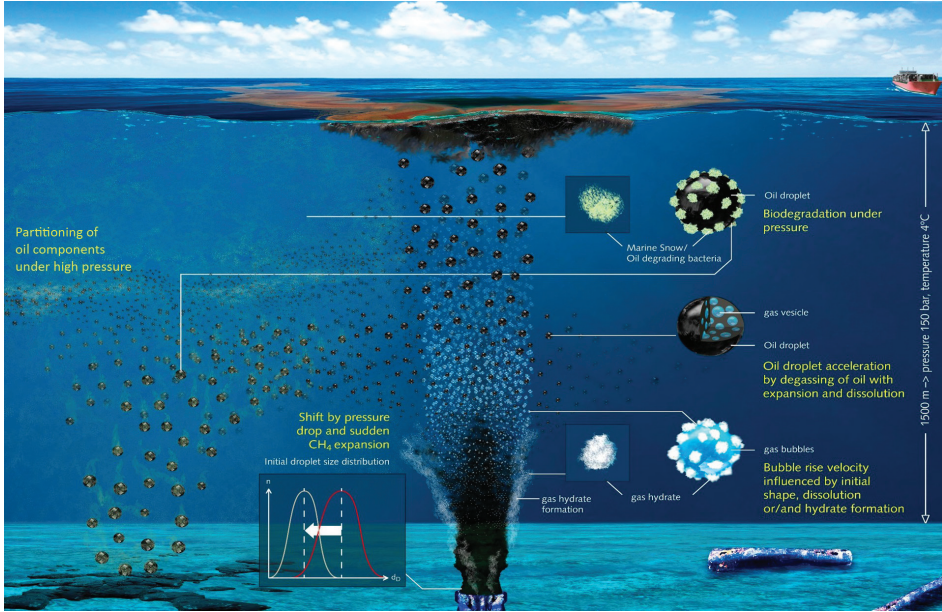


Fig. 2.2 Conceptual model sketch illustrating phenomena relevant to deep-sea oil spills.

$$Vi = \frac{We}{Re} \quad (2.8)$$

or the Ohnesorge number

$$Oh = \frac{\sqrt{We}}{Re} \quad (2.9)$$

can be used. The Weber number is a measure for the relative importance of the fluid's inertia in relation to the interfacial tension in two-phase flows. The Reynolds number is the ratio between inertial forces and viscous forces. The viscosity number and Ohnesorge number relate viscous forces to interfacial tension forces (and in case of the Ohnesorge number also to inertial forces) and are therefore stability indices for the dispersed phase fluid particles [Pes20a].

For the description of the rise behavior of fluid particles (see chapter 2.5.2), the particle Reynolds number

$$Re_p = \frac{u_p \cdot d_p \cdot \rho_c}{\eta_c} \quad (2.10)$$

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is required. As it describes the flow of the continuous phase around a (fluid) particle with the (relative) velocity  $u_p$ , the density  $\rho_c$  and viscosity  $\eta_c$  of the continuous phase are used. The particle diameter  $d_p$  serves as the characteristic length. The Eötvös number, also referred to as Bond number, relates gravitational forces to interfacial tension forces and is therefore important for buoyancy-driven processes. It is defined as

$$Eo = \frac{g \cdot \Delta\rho \cdot d_p^2}{\sigma}, \quad (2.11)$$

with the density difference  $\Delta\rho = \rho_c - \rho_d$  and the gravitational acceleration  $g$ . Together with the Eötvös number, the Morton number

$$Mo = \frac{g \cdot \eta_c^4 \cdot \Delta\rho}{\rho_c^2 \cdot \sigma^3} \quad (2.12)$$

characterizes the shape of the fluid particles by accounting for the physical properties of the substance system and the gravitational acceleration  $g$  [Pes20b]. Some correlations also use the Archimedes number

$$Ar = \frac{\rho_c \cdot \Delta\rho \cdot g \cdot d_p^3}{\eta_c^2}, \quad (2.13)$$

which can be interpreted as the ratio of buoyancy force to friction force, or the Best number

$$N_D = \frac{4 \cdot Ar}{3} \quad (2.14)$$

for the distinction of different validity regimes of the correlations and the calculation of the corresponding rise velocities [Cli78, Pes20b].

## Stages of Oil Spill Modeling

The modeling of deep-sea oil spills with regard to the dispersion, propagation, and distribution of oil (and gas) can be subdivided into four stages. Each of the stages addresses a specific set of questions and the stages sequence spatially and logically one after the other, from the rather small-scale investigation of the droplet breakup directly at the blowout site, through the modeling of the plume as well as individual drops in the water column, to the large-scale transport of the oil masses and their final distribution in the ocean.

**1. Oil dispersion models** focus on the droplet size distribution (DSD) that results from the blowout directly at the spill site. The predictive correlations yield the DSD or characteristic droplet diameters thereof. A fundamental work for the prediction of the maximum stable droplet size in liquid-liquid dispersions has been published by Hinze