Transfer, Interfaces and Processes (TIPS) – ULB

1. Experimental and/or numerical study of the vapour cloud surrounding an evaporating droplet

Summary: The evaporation rate of a deposited droplet is heavily influenced by the shape of the vapour cloud surrounding the droplet. At ULB-TIPS we have developed a powerful interferometric technique capable of measuring the vapour cloud surrounding such a droplet, for the first time. In parallel we are developing numerical codes with COMSOL Multiphysics to simulate the vapour cloud configuration, and calculate important quantities such as the total evaporation rate. Although apparently an academic subject, evaporative droplets are widely studied nowadays for various applications such as nanoparticle deposition, cleaning, or heat transfer technologies. The goal of the present thesis would be to master the experimental technique, perform the image treatment and compare the results to the performed numerical simulations.

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2. Evaporation of thin films of solutions and Bénard-Marangoni instability

Summary: When a thin layer of liquid dries by evaporation, the layer thickness decreases until its complete disappearance. During this process, a temperature gradient due to the latent heat of vaporization develops in the layer. This gradient can induce flow motion inside the liquid. Indeed, for small layer thickness, a flow in the form of hexagonal cells develops (Bénard-Marangoni instability) and the flow structure evolves with time. This kind of convective motion is well-known nowadays and we propose to extend the existing studies to the evaporation of two kinds of solutions: binary liquid mixtures and particle suspensions. What is the influence of a second component on the hydrodynamic flow and on the evaporation kinetics? The evaporation of a thin film of nanoparticle (or microparticle) suspension leads to the deposition of these particles on the surface. Is the deposit structure random or regularly organized such as hexagonal cells? Which parameters influence the pattern deposition? These questions are crucial for mastering deposition and coating techniques. In this experimental investigation, the flow motion will be visualized using shadowgraphy and/or infrared methods while pattern deposition will be characterized using a 3D microscope, techniques to which the student will be trained.

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3. Modelling of the hysteresis of evaporation-induced contact angles due to surface roughness

Summary: A contact line (CL, here a gas-liquid-solid triple line) presents fundamental difficulties for modelling, as in the framework of classical thermo-hydrodynamics it typically brings about various kinds of singularities. For their resolution, one has to account for microscopic, generally “non-classical” phenomena in a small CL vicinity. One the other hand, it is in such vicinity that contact angles, in particular evaporation-induced ones for volatile liquids, are established as apparent at macroscales. Nonetheless, the smooth
transition between the micro- and macroscales is further complexified by surface roughness, ignored in most studies. To bridge the gap here is one of the objectives of the ULB-TIPs department under the ongoing IAP and ESA projects and collaborations with Imperial College London (S. Kalliadasis) and Cardiff University (N. Savva), and a contribution to this study is hereby proposed as a MT subject. The roughness will first be modeled by simple modulations of the solid surface, such as a sinusoid or a bump/dimple of various amplitudes and wavelengths. The analysis will be carried out in the framework of the lubrication film equation to be solved numerically using Mathematica® and, if necessary, other commercial software (e.g. COMSOL). The primary focus is on the determination of contact angles as a function of the CL position and velocity, from where information on the expected contact angle hysteresis is to be extracted.

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4. Evaporation and wetting dynamics on textured surfaces

Summary: When a droplet is deposited on a surface, it can adopt various geometries depending on the physico-chemical properties of the surface, i.e. its surface energy and roughness. Whereas a droplet of water can wet a smooth substrate with an hydrophilic behaviour, it can acquire a non-wetting shape when this surface is textured. Recent studies have shown that it is now possible to control the surface properties in order to produce completely wetting or non-wetting surfaces, which has a large application potential in coating technologies. However, the interaction of droplet on such complex textured surfaces is still poorly understood. Therefore, we proposed to conduct a study to improve the understanding of the phenomenon of wetting of structured surfaces. More especially, we will focus on the dynamics of the contact line on these complex surfaces (with or without evaporation, in spreading or in recession). Chemically and/or geometrically structured surfaces, with regular and/or disordered patterns will be provided by partner universities (KULeuven and UMons), in the framework of an IAP project. This experimental investigation can be extended to numerical or theoretical approaches according to the wish of the candidate. The student will be trained to techniques such as interferometry and image processing, and possibly to COMSOL Multiphysics software.

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5. Evaporation and wetting dynamics of drops on flat surfaces

Summary: How does a drop behave when it contacts a flat surface? Immediately after contact, the drop begins to spread on the solid substrate, i.e. the triple line is advancing. In the case of perfect wetting, the drop continues to spreads on the surface until to form a thin layer. In the case of partial wetting however, the triple line advances until to reach a position of maximum wetting area. After that, if the liquid is volatile and the atmosphere is not saturated, the triple line recedes due to evaporation. We propose here to
experimentally investigate the spreading/evaporation of liquid drops on flat substrates. In order to perform this study, the student will have to be trained to and to use optical interferometry techniques allowing to follow the drop radius evolution and the drop contact angle. Today, two theories face each other for describing the spreading of drops on flat surfaces: the hydrodynamic theory (i.e. Navier-Stokes equations with suitable boundary conditions) and the kinetic molecular theory (accounting for the discrete nature of molecules). One goal of the study will be to compare the experiments with both these theories in order to validate or invalidate them. This work is part of a large IAP project and involves collaboration with UMons (Prof. Joël De Coninck).

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6. Modeling of the Leidenfrost effect

Summary: When a drop is placed on a surface at a temperature sufficiently above the saturation (boiling) temperature, the drop does not boil but rather levitates over a thin layer of its own vapor. This effect is known as the “Leidenfrost” effect. This often happens in the kitchen when some amount of water is released on a very hot cooktop. Even if this effect has been intensively investigated in the past, many questions remain open or under debate, in particular as far as the modeling of the phenomenon is concerned. The understanding and an accurate modeling of the Leidenfrost phenomenon are therefore crucial, such effect playing an important role in devices to be cooled or in new micro-milli-fluidic systems. Here, we propose a theoretical study of the Leidenfrost effect where several questions will be tackled: i) flow motion inside the drop; ii) determination of evaporation rate distribution; ii) prediction of the drop lifetime. The student will have to be trained to the COMSOL multiphysics software, and possibly to MATHEMATICA.

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7. Self-propulsion of Leidenfrost drops on a thermal gradient

Summary: When a drop is placed on a surface at a temperature sufficiently above the saturation (boiling) temperature, the drop does not boil but rather levitates over a thin layer of its own vapour. This effect is known as the “Leidenfrost” effect. This nearly frictionless state provides an extreme mobility to the drop, making it difficult to immobilize. Recently, some researchers took benefit from this mobility and discovered that Leidenfrost drops can self-propel when deposited on asymmetrically structured surfaces (called a ratchet). In this situation, such drops can move in a preferential direction with velocities of the order of 10 cm/s. Today, one can think to use this effect in new milli- or micro-fluidic systems, where droplets need to be manipulated quickly. The Leidenfrost effect has mainly two advantages compared with regular micro-fluidic systems. Firstly, there is no contact with the walls which can be a potential source of pollution. Secondly, the velocity of the liquid is rather high. We propose here to study another kind of self-propulsion, depositing such a drop on a hot surface with a thermal gradient. The drop should experience a thermal asymmetry leading to the self-propulsion of the drop. A theoretical modelling has been already developed. We consider here performing an experimental investigation allowing a comparison with the theoretical modelling and tackling several questions, such as the direction of the drop motion, the velocities that can be reached, etc ...

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8. Modelling of Leidenfrost droplet propulsion on ratchets

Summary: It is well known that a water droplet released over an extremely hot solid substrate generally does not touch the plate but rather “levitates” over a thin vapor film generated by its own evaporation. Although discovered in 1756 and widely studied in connection with heat transfer technologies, this so-called Leidenfrost effect is the subject of a renewed interest nowadays, particularly in view of new perspectives in the field of microfluidics. Indeed, as the relatively small thermal conductivity of the vapor layer slows down the phase change process, while its low viscosity confers an extreme mobility to the drop, the control and manipulation of Leidenfrost drops proves to be quite handy and practicable in various ways. This is one of the objectives of the ULB-TIPs department under the ongoing IAP project in collaboration with Université de Liège (S. Dorbolo). More concretely, as a subject of the present MT, we propose a theoretical study on setting Leidenfrost droplets in motion, and hence transporting them between different locations, by means of ratchets (periodic modulations of the solid substrate surface). The modeling will include numerical resolution of the lubrication film equation for the vapor film by means of Mathematica® and, if necessary, other commercial software to which the student will be trained.

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9. Experimental study on the influence of an electrical field on the shape of sessile droplets

Summary: When applying an electric field on a droplet deposited on a conducting substrate, it has been found in literature that the shape of the drop becomes deformed. Increasing the potential of this field even further, the droplet ultimately reaches a cone-like shape (the Taylor cone) and a spray of small droplets can even be generated (an electro-spray, used for instance to deliver aerosols). Although this is a rather old problem (+- 1960), some questions still remain on the precise influence of the electric field on the shape and contact angle of the drop. On the other hand, when placed between two plates and applying a voltage, the droplet will exert a force to obtain its new equilibrium shape. The goal of the present thesis then is twofold. First, we would measure a full 3D shape of the droplet with digital interferometry as a function of the strength of the electric field in order to quantify the change in contact angle. The second goal is to measure the force in a wedge geometry precisely and examine its potential to use this configuration as a micro-actuator.

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10. Numerical study by averaged approach of gas-liquid meniscus motion in Hele-Shaw cell with chemically heterogeneous walls

Summary:
The study of fluid dynamics in confined geometries is a rapidly growing research field, because of the recent advances in micro-fluidic reactor development. The characteristic length scale of these devices is such that the physico-chemical properties of their surfaces are essential for the dynamics of liquid flowing inside. This work deals with the numerical simulation of a gas-liquid meniscus forced to flow in a Hele-Shaw cell (thin gap between two glass walls) with chemically heterogeneous walls (inducing a heterogeneous wettability), thanks to a model developed in partnership with Imperial College London. This model is based upon an averaged Cahn-Hilliard phase field equation and is simulated with the COMSOL Multiphysics software. After a training to the use of COMSOL, the goal of this master thesis will be to investigate the statistical
characteristics (correlation length, scale invariance, avalanche dynamics ...) of the gas-liquid contact line and to correlate them with the statistical parameters of the heterogeneities (size, standard deviation ...).

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11. Numerical study of the bubble growth on a heated plate coupled with a Marangoni flow

Summary: Evaporation takes place in numerous devices but its dynamics may be quite complex and its rigorous modelling is still partial since several phenomena are coupled. The improvement of devices using evaporation requires a better understanding of the coupling and the numerical simulation is an appropriate analysis tool. In this work, the growth of a gas bubble on a heated plate is investigated. The surrounding liquid contains dissolved gas and then the bubble grows by the combination of the liquid evaporation and the dissolved gas desorption, induced by the heat flux. Since there is a temperature gradient along the bubble interface, a Marangoni flow is triggered, which influences the mass and heat transports. A mathematical model, coupling momentum, heat and concentration transport, has been developed in partnership with the Institut de Mécanique des Fluides de Toulouse (Profs C. Colin and D. Legendre). It is simulated with the COMSOL Multiphysics software. The goal of this master thesis is to investigate the influence of the Marangoni flow on the heat and mass transfer rates and to determine the influence of the operating (e.g. heat flux) and physico-chemical (e.g. viscosity) parameters on the bubble growth dynamics.

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12. Contribution to the study of CO₂ absorption dynamics in monoethanolamine aqueous solution by Mach-Zehnder interferometry

Summary: The study of processes enabling the CO₂ capture from flue gas is an important research field for chemical engineers in the framework of reduction of greenhouse gas emission. The Mach-Zehnder interferometer (MZI) is an experimental device enabling the visualization of CO₂ absorption in liquid at the interfacial scale. However, we see only the global effect of several phenomena and thus the results interpretation may be difficult. The goal of this work is to study the evolution of optical characteristics of aqueous solution of monoethanolamine (MEA) with CO₂ absorption. It will provide a comprehensive framework, which will enable correlating mathematical models based upon the fundamental physico-chemical phenomena with the results experimentally obtained by MZI. The software COMSOL Multiphysics and MATLAB will be used and calibration curves will be determined by refractometry. A comparison between model and experiments will be eventually realized.

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13. Experimental study of interfacial turbulence in evaporating liquids

Summary: When a liquid layer evaporates into air, the liquid-air interface is cooled by the consumption of latent heat of vaporization. A gradient of temperature then occurs, which can lead to a symmetry-breaking hydrodynamic instability due to the surface tension variation with temperature (the so-called Marangoni effect). This Bénard-Marangoni instability manifests itself in the form of hexagonal convection cells, with the flow rising in the middle of each hexagon (warmer), and descending at its periphery (colder). It is also well-known that these hexagonal structures may contain defects such as pentagon-heptagon pairs or chains, especially when the rate of cooling (due to evaporation) becomes larger. At even larger evaporation rates, the dynamics is more complex, with convection cells spontaneously splitting into smaller ones, and other cells simply collapsing. This fascinating chaotic dynamics has not yet been studied in details, despite the interest this has for heat transfer applications and micro-mixing, in addition with nano- (or micro-)particle deposition. A lot of infrared thermography data has been acquired in the TIPs laboratory during recent years. The goal of this MT will be to: i) develop image processing routines allowing to extract useful quantities from the acquired images; ii) to carry out new experiments using an infrared camera; iii) to analyze the dynamics of defects in the structures and of events such as splitting and coalescence of convective cells, in order to understand this “interfacial turbulence”. This will be carried out in collaboration with Univ. of Berkeley (Prof. E. Knobloch).

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14. Numerical analysis of volatile binary droplets interactions mediated by the gas phase

Summary: Droplets of volatile liquids deposited on a flat plate may have surprisingly complex dynamics. Very recently, a team from Stanford University has published a paper in Nature [N. J. Cira, A. Benusiglio & M. Prakash, “Vapour-mediated sensing and motility in two-component droplets”, 2015] showing that droplets can be attracted to each other when they are sufficiently far apart, while they may repel each other when they are too close. This behaviour, which evokes the interaction forces between usual molecules, can result in very complex dynamical regimes (“dancing droplets”, see https://www.youtube.com/watch?v=K8Wx2PHIYGI), even though the droplets are here millimetric in size. Both the long-range attraction and the short-range repulsion can be explained by the effect of surface tension variations with concentration of the volatile component in the droplets, but the long-range attraction also involves vapour exchange between droplets, via the gas phase). In addition with their fascinating character, such spontaneously moving droplets might have very promising applications in microfluidics, a rapidly expanding technological field aiming to use droplets as micro-reactors within which chemical analysis or synthesis can be accomplished very fast, with small amounts of liquids, and in a parallel manner. The goal of this MT will be to numerically investigate the dynamics of binary droplets interacting via the gas phase by vapour exchange, on the basis of a model recently developed in TIPs. The student will be trained to various simulation software (such as e.g. COMSOL Multiphysics, Mathematica, ...) and will develop a numerical code allowing to solve a system of partial differential equations
15. Study of falling liquid film stability with negative gravity

**Description:** A film of condensation that forms on a cold ceiling is unstable with respect to gravity (called negative gravity) and breaks down into a myriad of regularly spaced droplets, the distance of which can be predicted by linear stability analysis; this is referred to the well-known Rayleigh-Taylor instability. In the case of a slight inclination, the film flows in the direction of the slope and the symmetry is broken. Another type of instability can arise in this case, which produces travelling wave structures, like on a windshield of a car under rain fall; this is referred to the hydrodynamic instability. The coupling of the two above-mentioned instabilities has never been studied, despite the fact that it is of great interest, not only on the fundamental point of view, but also on a practical point of view. Indeed, many industrial processes involving falling films (evaporators, cooling systems, bioreactors) have inclined surfaces where wave structures play a crucial role, not only in the stability of the film but also in the heat and mass transfer processes. The objective of this project will be from a dedicated low-dimension theoretical model, to determine the phase diagram of such a complex system, using linear and non-linear analytical analysis, together with numerical simulations. The model will then be validated with experimental data obtained from an in-house experimental set-up.

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16. Contribution to the development of a continuous microfluidic process for the crystallization of pharmaceutical compounds (Collaboration : UCB (LEGOMEDIC project) )

**Description:** As the latest stage in the continuous production of active pharmaceutical ingredients (API), the crystallization step further purifies the API and introduces the desired crystalline morphology combined with a suited crystal size distribution. Microfluidics can offer here a wide range of new tools that permits to revisit the formation of crystals in solution and yields insights into crystallization processes. As such existing industrial processes also benefit for instance from this newly acquired thermodynamic and kinetic data and shed a new light on nucleation and growth mechanisms. A microfluidic chip allows fine-tuning of control parameters for crystallisation, such as residence time, concentration, temperature constraint and Reynolds number. A special attention will therefore be paid in this project to the influence of the flow and residence time (Reynolds number and channel topology) on the nucleation kinetics, since to our knowledge, no systematic study exists on that topic and because it has an important impact on the selectivity conditions for polymorphs, which are crucial in the production of pharmaceutical compounds.
In this project a microfluidic chip suited for crystallization will be developed, which consists in designing and fabricating the microfluidic chip by soft lithography and using high sensitive microscopy for characterization. This chip should allow determining the crystal solubility, controlling the shear stress from various flow structures and determining its influence on the germination kinetics and the morphological selection. The experimental study will be complemented with direct numerical simulations using dedicated CFD (Computational Fluid Dynamics) software’s. At all times, the direct comparison with the existing industrial processes will be made in order to check the beneficial influence of the microfluidic work.

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Type of work: a whole range of theoretical, computational and experimental work

17. Contribution to the development of a microfluidic distillation for the purification and concentration of hydrogen peroxide (Collaboration: Solvay (MICROECO project))

Description: Production of hydrogen peroxide on-site by direct synthesis (from hydrogen, H\textsubscript{2} and oxygen, O\textsubscript{2}) is favoured because of its sustainability (elimination of by-products from traditional synthesis of H\textsubscript{2}O\textsubscript{2} and reduction of CO\textsubscript{2} emission due to transportation). Due to the hazardous character of direct synthesis of H\textsubscript{2}O\textsubscript{2}, the process cannot be performed in bulk. Continuous production in microfluidic channel solves this issue. Production of H\textsubscript{2}O\textsubscript{2} by direct synthesis requires dissolution of hydrogen and oxygen in a solvent in which in presence of catalyst synthesis occurs. Methanol (MeOH) is preferred as solvent because it dissolves gases in higher amount than H\textsubscript{2}O and the catalyst efficiency is increased. After production, the solvent has to be removed from the product H\textsubscript{2}O\textsubscript{2} and by-product H\textsubscript{2}O which can be done by pervaporation in microfluidics. Some aspects of the ternary system (H\textsubscript{2}O\textsubscript{2}/H\textsubscript{2}O/MeOH) membrane pervaporation in microfluidic were already investigated in our group.

![Diagram of micro-distillation device](image)

Fig. 1. Scheme of the micro-distillation device: (a) general overview, (b) side view

We would like to investigate further the influence of porosity and thickness of the membrane on the efficiency of distillation.

This project will require the preparation of different porosity of PDMS membranes and building the microchip as well as operation of chip for distillation of ternary mixture.

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18. **Surface rheology using microfluidic-based antibubbles** (Collaboration: PAI (Micromast project))

**Description**: Although the concepts of surface viscosity and elasticity date back to the Belgian scientist Plateau in the 19th century, they have only later been formalized mathematically and subsequently been used in quantitative descriptions of surface flows. The concepts of surface viscosity and elasticity (Marangoni) effects are intimately related, and are not always distinguishable in an experimental setting. One such setting is the antibubble. An antibubble is a spherical object that can be seen as the opposite of a soap bubble. It consists in a spherical air film surrounded in and outside by liquid mixture full of surfactant. Yet, hydrostatic pressure will drain the air film to the north pole making the antibubble unstable. A recent model developed by the promoter allows rationalizing the experimental data based on the sole effect of surface rheology.

At the moment, antibubbles are generated by pouring liquid mixture inside a beaker by hand which lead discrepancies in size, initial gas volume,... from experiments to experiments. The project will thus consist in designing a tool to create antibubbles of calibrated diameter. Then by studying the mean lifetimes of antibubbles, this system will thus act as a surface rheometer, determining the surface viscoelasticity of a fluid/fluid interface. Since lifetimes obey to an exponential distribution, there is a need to generate a large number of antibubbles in order to obtain statistically reliable mean lifetimes.

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**Type of work**: Theoretical/Experimental


**Other subjects with details on demand**:

(With the possibility of doing a 3-months training in the company)

- Investigation of rupture of glass fibers during their fabrication process
  (Industrial partner: 3B)
- Study of edge instability occurring during the slot dye coating of an adhesive
  (Industrial partner: MACTAC)
- Contribution to the development of a microfluidic device for the production of micrometer beads for protein purification
  (Industrial partner: Mecasoft, UCB)

**Promotor**: Benoit Scheid

**Type de work**: Theoretical/Experimental

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