Physiochemical Hydrodynamics of Droplets and Bubbles Far from Equilibrium

Detlef Lohse

2018 Balzan Prize for Fluid Dynamics

Balzan GPC Advisers: Étienne Ghys, Carlo Wyss
Deputy Supervisors: Sander Huisman, Assistant Professor, Physics of Fluids group, University of Twente; Chao Sun and Xuehua Zhang, Part-time Professors, Physics of Fluids group, University of Twente
Affiliated Institutions: Physics of Fluids (PoF) Group, Max Planck Center Twente for Complex Fluid Dynamics, JM Burgers Center for Fluid Dynamics, Mesa+ Institute, Department of Science and Technology, University of Twente; Max Planck Institute for Dynamics and Self-Organization, Göttingen, Germany
Period: 2018-

Detlef Lohse is Chair Holder at the Physics of Fluids group at the University of Twente, The Netherlands.

The objectives of his research project are: to explore well-defined examples of multi-component and multi-phase liquid systems involving droplets and bubbles, for which a successful quantitative description and one-to-one comparison between well-controlled experiments, theory, and numerics can potentially be achieved; to identify the underlying principles; and to place these examples in the context of relevant applications of multi-component and multi-phase liquid systems. In particular, the team will focus on solvent exchange and multi-component droplet nucleation and dissolution on a small scale, and on multi-phase and multi-component turbulent Taylor-Couette flow on a large scale.

Background

Classical hydrodynamics focuses on pure liquids. However, in most cases in nature and technology, liquid systems are multi-component and multi-phase, with phase transitions between the phases, thus far from equilibrium. This state of non-equilibrium can be driven by flow, mixture, phase transitions, chemical reactions, electrical current, heat, etc. In the 1960s, Veniamin Levich dealt with such systems in *Physiochemical Hydrodynamics*, where the theoretical and mathematical concepts of the field are described. Today, the scientific community, and the field of physiochemical hydrodynamics in particular, has witnessed tremendous development, making the experimental, instrumental, and numerical means to deal with the problems more widely available.

These developments are more than timely, as the relevance of the physiochemical hydrodynamics of multi-component and multi-phase liquids has become increasingly important in light of the challenges mankind faces in the twenty-first century, especially as regards energy: storage, batteries, hydrogen production by electrolysis, CO2 capture, polymeric solar cell manufacturing, biofuel production, and catalysis. Other challenges are presented by health and medical issues, like diagnostics or the production and purification of drugs; by environmental issues like flotation, water purification, membrane management, and separation technology; by food processing and food safety issues; or by issues in modern production technologies like additive manufacturing, inkjet printing, and the paint and coatings industry.

Traditionally, these challenges have been approached from the point of view of pure engineering, and less in the spirit of a scientist/engineer like Levich. On the other hand, classical hydrodynamics has focused on pure and single-phase liquids. In recent years, however, the scientific community has begun to bridge the gap between fluid dynamics, chemical engineering and colloid and interface science, with the aim of mastering and better controlling multi-component and multi-phase fluid dynamics systems far from equilibrium by coming to a quantitative understanding of them. To achieve this aim, controlled experiments and numerical simulations for idealized setups must be performed, allowing for a one-to-one comparison of experiments and numerics/theory that will test the theoretical understanding. Thus, taking Levich's *Physiochemical Hydrodynamics* as a point of departure, the field has built on and benefited from the developments of modern microfluidics, microfabrication, digital (high-speed) imaging technology, confocal microscopy, atomic force microscopy, and various computational techniques and opportunities for high-performance computing, thus ushering in a golden age of fluid dynamics.

Lohse's Physics of Fluids group started to work in the direction of the physiochemical hydrodynamics of multi-component systems only a few years ago. Based on their initial work on surface nanobubbles and nanodroplets, the team looked into droplet nucleation in solvent exchange processes, including purely theoretical and numerical studies, as well as the evaporation of multi-component droplets and the diffusive and inertial dynamics of so-called plasmonic bubbles, for which phase transitions are crucial. Two important examples of the team's research can be seen in their small-scale studies on the impact of droplets on superheated surfaces (Leidenfrost effect) as well as their large-scale work on thermal convection around the boiling point of liquids (Nusselt number, turbulent Rayleigh-Bénard convection).

Objectives

Lohse's team will focus on the physiochemical hydrodynamics of (multi-component) droplets and bubbles far from equilibrium, exploring examples of such systems for which a successful quantitative description and one-to-one comparison between well-controlled experiments and theory and numerics can be achieved. They also aim to identify the underlying principles, and to put these examples in a context of relevant applications of multi-component and multi-phase liquid systems. The plan is to finance a postdoctoral position with the Balzan funds. The chosen researcher will be part of Lohse's Physics of Fluids research group, and thus will benefit from its intellectual and technological infrastructure.

Research Platforms and Programme

The two main experimental platforms – one small-scale and one large-scale – will address the physiochemical hydrodynamics of multi-component droplet and bubble systems far from equilibrium. The small-scale project will involve a solvent-exchange microchannel, and the large-scale project will use the Taylor-Couette apparatus as the platform for multi-component-multi-phase experiments.

Small Scale Experiments: Multi-Component Droplets with Phase Transitions

Controlled drop nucleation and growth experiments can be performed through the so-called solvent exchange process. Lohse's team will show how an oil-saturated ethanol solution with relatively high oil solubility is pushed away by an oil-saturated water solution with relatively low oil solubility. In the mixing zone, a temporal oil oversaturation will emerge, leading to droplet nucleation and growth on the substrate.

Lohse's group will study how this growth process depends on flow velocity, channel geometry and scale, oil types (allowing for multiple oils in one experiment), oil saturations, surface properties, and neighbouring droplets.

In particular, the team plans to work on multi-component systems, with several oils of different solubilities dissolved in the aqueous phase, with the aim of trying to determine relative solubilities and calculate this a priori. They also plan to sequentially push different solutions through the microchannel, so that concentration gradients will evolve in the emerging droplets. In general, these will cause flow within the droplet, which may interact with the outside flow. When pushing clean water through the channel, the multi-component droplets will selectively dissolve, creating even more methods for concentrating or diluting certain components.

Experimental methods to be employed include high-speed imaging, particle image velocimetry, confocal microscopy and diagnostics techniques to measure the composition of the droplets. The substrate can be chemically patterned to fix the positions of the droplet nucleation. In addition to the experiments and analytical calculations for the diffusive processes around single surface bubbles and droplets, numerical simulations for controlled populations of nanobubbles/nanodroplets will also be performed by numerically solving the advection-diffusion equations with the respective boundary conditions. The methods will be direct finite difference simulations of the continuum equations with immersed boundary methods (IBM) and MD simulations. The team is very optimistic about achieving a favourable one-to-one comparison between experiment, theory, and numerical simulations, at least for the controlled conditions of chemically or geometrically pre-patterned surfaces.

Large-Scale Experiments: Multi-Phase and Multi-Component Turbulence

For large-scale experiments, the platform will be the Taylor-Couette system, i.e., two coaxial co- or counter-rotating cylinders with a fluid in between. Up to now, Lohse's team has used the Taylor-Couette system only for single-phase flow and for bubbly flow, but they are optimistic that, in spite of the complexity of turbulent multi-phase flow with phase transitions, they will be able to perform controlled experiments in their new Twente turbulent Taylor-Couette setup, which has excellent temperature control and which allows very precise measurements of the overall drag. Moreover, it can be operated with low-temperature boiling liquids. At phase transition close to the boiling point, they expect the vapour bubbles to emerge in the low-pressure regions, namely within the so-called Taylor vortices. This will have major consequences for the local velocity and temperature fields, the overall flow organization, and the drag laws. In addition to the Reynolds numbers of the inner and outer cylinder, the crucial control parameter is the liquid temperature and its thermal properties. For strong turbulence, vapour bubbles will form in the Taylor vortices, affecting the overall drag, which can be measured by adjusting the required torque to rotate the cylinders at fixed velocity. Lohse's team would thus be the first to perform controlled drag measurements in a turbulent boiling liquid.

Along with the global measurements of the drag, the team will also make local optical high-speed observations of vapour bubble dynamics and densities, to be measured with 3D particle tracking velocimetry (PTV), and the velocity field, to be measured with 3D particle image velocimetry (PIV).

A second case to be analysed is the strongly turbulent flow of (multi-component) oil and water, also in the Taylor-Couette geometry. In regimes that are strongly turbulent (i.e., far from equilibrium) and without surfactants (e.g., used to stabilize mayonnaise), the dynamics of the oil-water mixture is totally unexplored. In attempting to answer questions as to whether the inversion is hysteretic or not, or the extent to which the overall viscosity depends on the oil concentration, or what the typical droplet size and size distribution is in both cases, the team wants to go beyond two-component mixtures by analysing a ternary system with three components like the famous ouzo system (oil, ethanol, water). As in the small-scale experiments, solvent exchange will be performed, but in this case in the turbulent Taylor-Couette system.

The backbone of the numerical simulations in this part of the project will be a finite difference code with which the advection-diffusion equations for the temperature and the gas concentrations of the various reactants and reaction products will be simulated, fully coupled to the Navier-Stokes equation. This massively parallel central-finite-difference code has been developed and optimized within Lohse's group, and applied to highly turbulent Taylor-Couette and Rayleigh-Bénard flows. Thanks to the combination of open MP and MPI parallelization directives, the method can efficiently run on tens of thousands of processors, thus tackling unprecedentely high Reynolds number turbulent flows. Lohse's team has recently improved their scheme through a multiple resolution strategy that enables them to deal with potentially different time scales set by the scalar field and the velocity field.

The emerging and growing or shrinking droplets and bubbles will be represented with the immersed boundary (IB) method, which makes it possible to implement the effect of the diffusive interaction of bubbles and droplets in general and of Ostwald ripening in particular, by adjusting the surface concentration at the bubble-liquid or drop-liquid interface according to Henry's Law. The team is able to employ these methods for multi-component systems, emphasizing that the coupling of IB and FD for bubbles or droplets and strongly turbulent flow is a major new development which they wish to push ahead with this project.

Presentation of Results

In the last year of the project, Lohse as project director will organize an international Balzan workshop, "Physicochemical Hydrodynamics of Droplets and Bubbles Far from Equilibrium," at the Lorentz Center in Leiden. This will give the Balzan Foundation, the project, and the subject area greater visibility. It will be an excellent opportunity to exchange views on a relatively narrow, well-defined subject, in an international group of about fifty scientists.