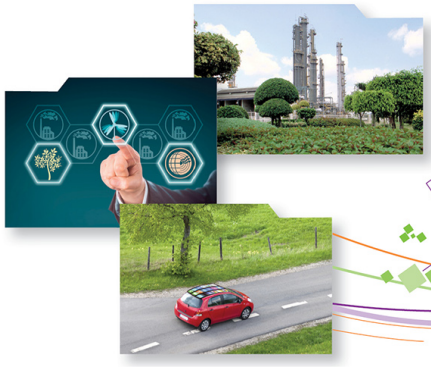


Science@ifpen
Issue 49 - October 2022

Written on 30 November 2022

15 minutes of reading

News

Fundamental Research

The context of climate change and energy demand requires an acceleration in R&I efforts **to develop the low-carbon technologies of the energy and ecological transition.**

These developments necessitate multidisciplinary innovative research combining pragmatism with industrial realities and environmental constraints.

Research conducted within the Applied Physical Chemistry and Mechanics Division meets the need for a cross-cutting scientific approach.

By drawing on the complementary disciplines of its five departments, the division is able to offer several different ways of representing phenomena.

Irrespective of the applications considered, it is necessary to establish a dialog between different physics - and to do so on different scales - in order to explore, understand, characterize and model the behavior of materials and structures in their environment or that of complex fluids at the source of numerous phenomena involved in processes.

The ambitious objective we are pursuing is **to connect the discrete and molecular world of physical chemistry to the continuous and more macroscopic world of mechanics and thermodynamics**. To achieve this we draw on a pool of experimental and numerical tools, appropriate for the various study scales, and a solid bedrock of scientific knowledge, perpetually enriched by academic and industrial partnerships and via participation in collaborative projects.

We are delighted to give you the opportunity to explore the broad lines of this research through a few recent examples illustrating the four priority areas of our scientific policy: complex fluids, fluid/system interactions, electrochemical systems and structures in their environment.

We hope you enjoy reading these articles.



Thierry Bécue
Director, IFPEN Applied Physical Chemistry and Mechanics Division



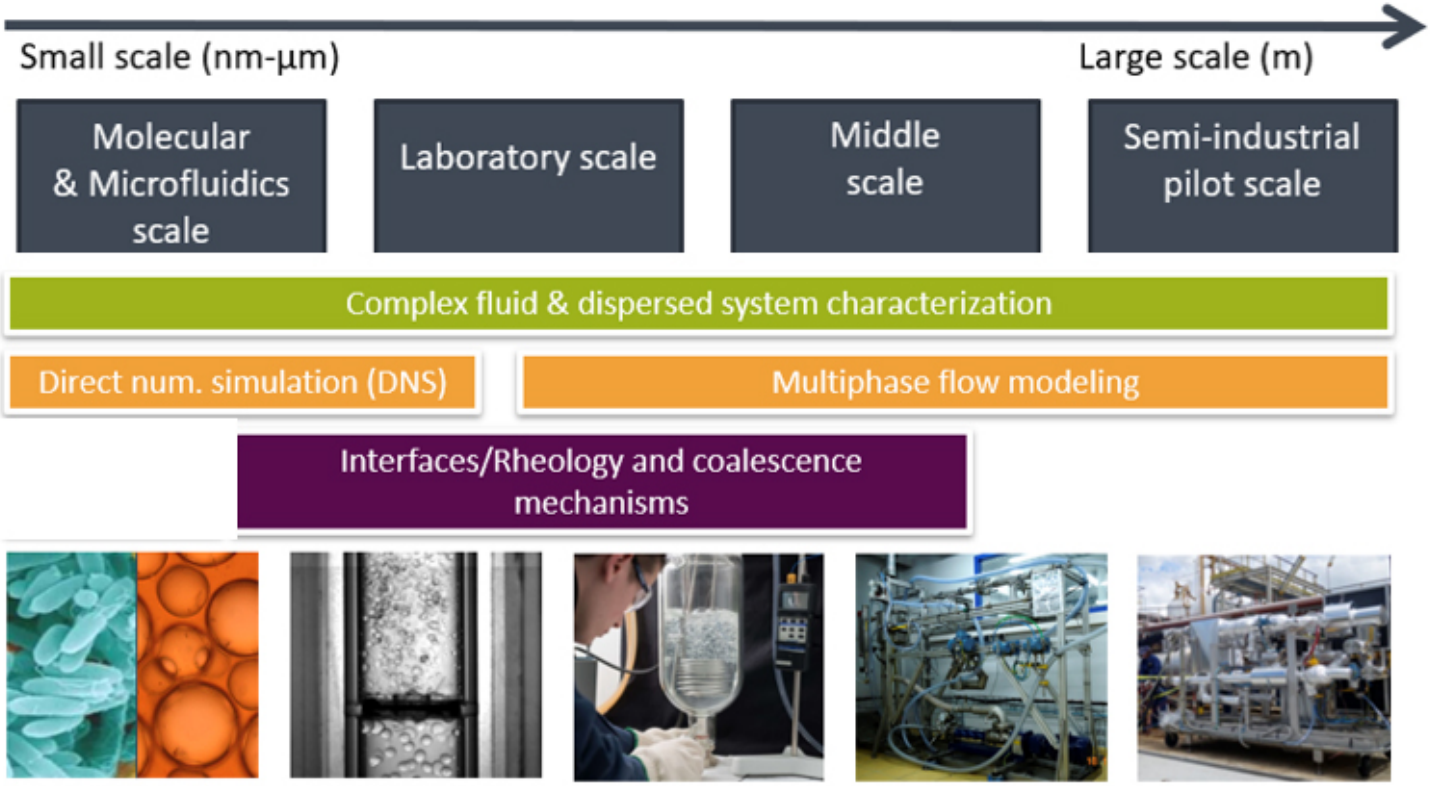
Laurent Cangémi
Deputy Scientific Director, IFPEN Applied Physical Chemistry and Mechanics Division

LES BRÈVES

Numerous IFPEN applications, from liquid-liquid extraction for biofuel production to the flotation process for the separation of microplastics, bring into play interactions between a fluid and the large number of inclusions (bubbles, drops) it contains.

The difficulty involved in describing these interactions is related to the flows as such but also to the physical phenomena taking place on various scales: from colloidal forces acting on a molecular level through to hydrodynamic forces acting at flow level.

These forces are not independent. The physics of the interactions at the local level of the interface between the two phases (fluid and inclusions) govern the macroscopic behavior of the flow, via rupture/coalescence phenomena, and hence inclusion size as well. In turn, this size influences the hydrodynamic forces the inclusions are subject to.



No generally accepted theory exists to describe all of these phenomena. As a result, each new case study requires the performance of numerical simulations or experimental data acquisition. Molecular simulations and precise experiments make it possible to trace the interfacial properties of interest, essential for a precise description of rupture/coalescence phenomena.

Recent research of this type made it possible to trace the properties of the liquid-liquid and liquid-gas interfaces along which amphiphilic polymers[a] had been distributed [1]. In parallel, purely hydrodynamic simulations gave access to drainage times in imposed force liquid-liquid configurations [2]. The combined used of these two approaches is very promising in terms of obtaining the critical

thickness for which the film breaks, with a view to injecting it into drainage time scaling laws. Moreover, it will lead to better determination of fluid and interface rheology, with the aim of direct integration in hydrodynamic codes, via constitutive laws.

An additional step to feed into engineering models (Euler-Euler-type) will be to consider a large number of coalescent inclusions and “average” their dynamics. In order to dimension industrial processes, it will then be possible to no longer consider each of the inclusions with its own movement in numerical simulations but, instead, an “averaged” fluid, leading to significant calculation time savings. The shift away from the models previously used towards this type of averaged approach is the focus of a research project conducted as part of a collaboration[b] with the CEA.

a- Having both hydrophilic and hydrophobic parts
b- Also involving IFPEN’s “Process Design and Modeling” division

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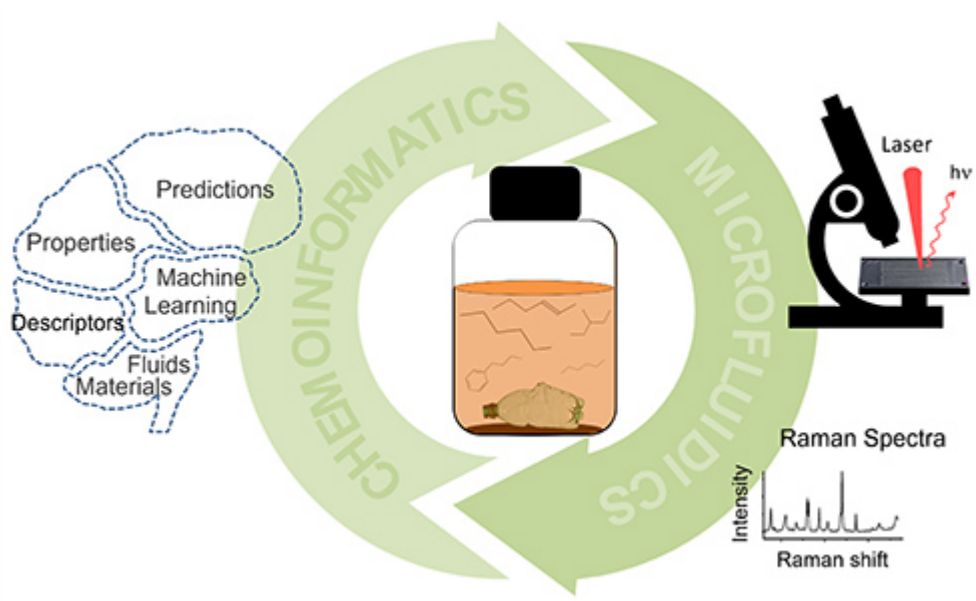
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Coalescence and interfacial property: from the microscopic to the macroscopic



For many industrial applications, such as the chemical recycling of plastics, or in order to ensure compatibility between polymers and new fuels, it is essential to anticipate interactions between materials and fluids. Bringing them into contact with each other can trigger ageing phenomena resulting in a loss of their initial performances. It is thus crucial to have a better understanding of these phenomena and the subsequent degradation kinetics in order to be able to design equipment as precisely as possible.

The ageing of materials in the presence of a fluid can be evaluated by measuring the mass of samples of a few cubic centimeters immersed in the fluids concerned over variable periods of time. The major drawbacks with this method are that it requires extremely long and expensive experimental campaigns and it is poorly adapted to some specific situations: sensitive products or conditions (toxicity, potential dangers, extreme temperature and pressure conditions, etc.). To overcome this problem, two possible options are being rolled out together at IFPEN: the development of predictive models and the reduction of experimental scale.

The first option involves turning to Chemoinformatics, one aspect of which is the use of artificial intelligence to predict properties on the basis of reference data [1]. Its application has already made it possible to model the quantity of fluid penetrating a polymer during prolonged contact between the two [2]. In order to increase the number of potential applications of these models (new polymer/fluid pairings, other temperature and pressure ranges, etc.) or make them more robust, it is now important to supplement existing data via the large-scale and targeted acquisition of new experimental results. To achieve this, new techniques need to be considered.

To this end, the field of Microfluidics broadens the spectrum of experimental options. This technique consists in studying the behavior of fluids and/or materials in miniaturized systems, such as chips, containing channels a few tens of micrometers in diameter (Figure 1). This scale reduction offers considerable advantages in terms of cost, experimental safety and time. The small volumes involved reduce the quantities of matter required and limit operating risks. Moreover, the method is naturally suitable for “high-throughput” experimental data production (parallelization, acceleration via surface/volume ratio increase). In addition, combining non-destructive methods, such as Raman

spectroscopy, makes it possible to study phenomena in situ, including in harsh conditions, due to the fact that these chips are able to function in high-pressure, high-temperature conditions [3].

All these aspects make the combined use of Chemoinformatics and Microfluidics for studying material-fluid interactions an obvious solution. More broadly, their combination opens up interesting new avenues for the development of robust models, i.e., based on numerous and relevant data, and capable of also taking into account new compounds as well as new pressure and temperature conditions.

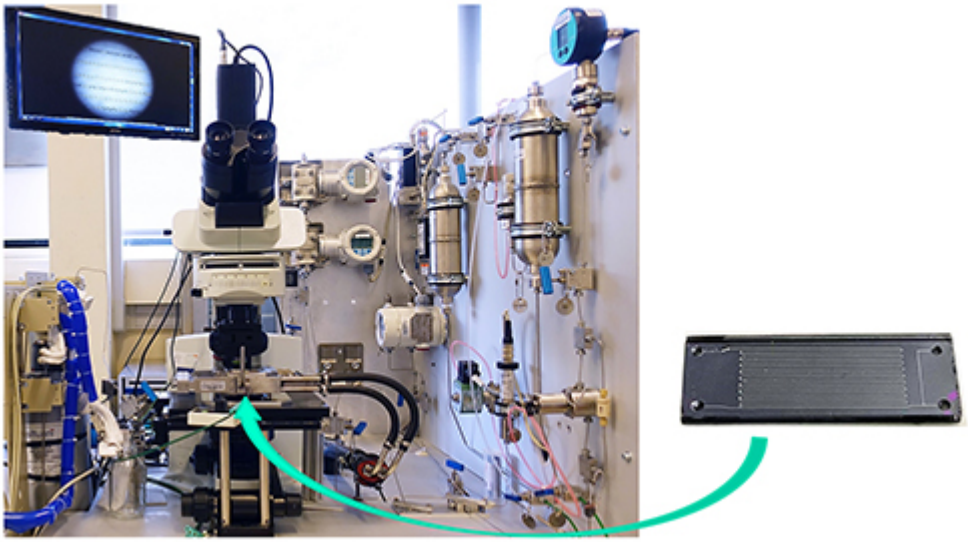


Figure 1. High-pressure, high-temperature (100 bar – 300°C) experimental set-up (left) and microfluidic chip (right)

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Microfluidics and Chemoinformatics: a highly complementary approach to studying material/fluid compatibility

Supported by its recognized experience in the development of energy production solutions, IFPEN has acquired in-depth expertise in the field of materials for functional use. Today, this expertise is being harnessed to address the emerging challenges of the energy transition, from new battery systems through to the thermal and energy management of processes (compressed air energy storage, process decarbonization, etc.) and new types of powertrain.

Functionalizing a material - i.e., giving it controlled properties as a function of its structure, formulation or chemistry - requires the implementation of approaches combining different physics and different scales.

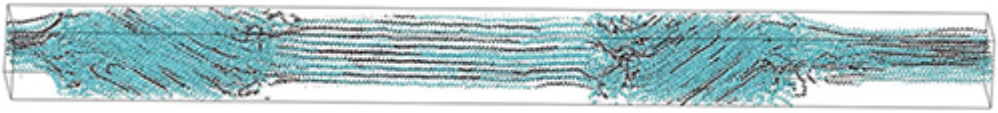
Considering the growth in computing power available for numerical simulation, over recent years IFPEN's teams have been implementing a global multi-scale modeling approach, aimed at bridging the gap between the understanding of mechanisms on a small scale (molecular or microstructural) and macroscopic, thermodynamic and mechanical approaches [1-4]. This global approach is hinged around two complementary areas:

- Molecular simulation for an understanding of the physicochemical phenomena or detailed coupling with mechanics.
- The simulation of functional usage microstructures making it possible to optimize mass transfer and thermal and mechanical properties within "controlled" architecture materials, for new solutions based on 3D printing.

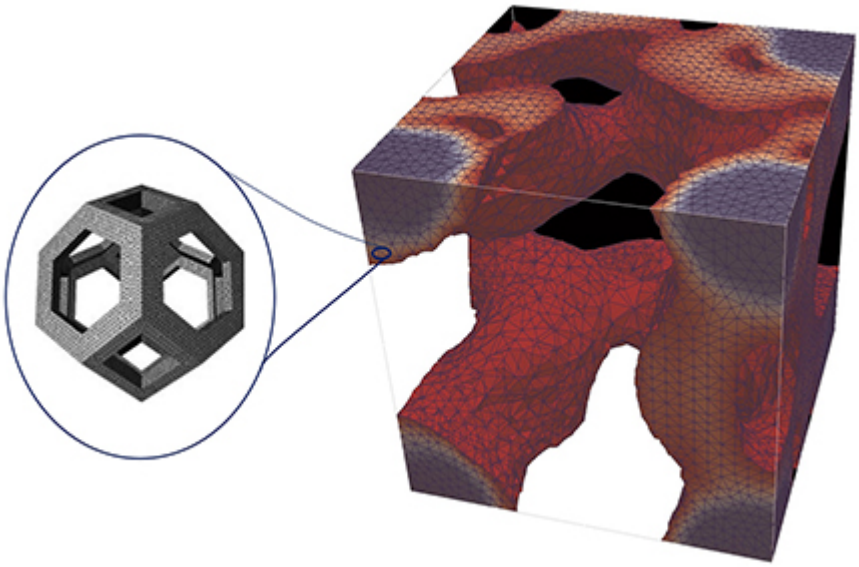
The approach was initially used to predict, from the molecular scale, the elastic properties of materials presenting several characteristic scales, such as catalyst supports or semi-crystalline polymers (figure 1. a) [1], [2] and [4].

This methodology will also make it possible to improve the prediction of physicochemical properties, such as species solubility in a polymer material and the understanding of interactions with certain solvents (plastic recycling), the understanding of species diffusion in confined media (zeolites, membranes, etc.), or, on a larger scale, to build new structured materials via topological optimization for electric motors, heat exchangers (Figure 1.b) or storage systems [3].

Figure 1. Examples of microstructures and physical problems on different scales.



a) Molecular amorphous/crystal assembly in a semi-crystalline polymer subjected to tensile stress with the appearance of cavitation [1]



b) Porous microstructure (left) and variable density mesostructure (right) obtained via topological optimization of a thermo-poro-mechanical problem [3]

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Materials for energy, a scale transition for an energy transition

The field of electrolyte thermodynamics is a strategic one for IFPEN since it contributes to a number of its existing technological innovations, as well as those currently being developed.

Examples of applications include:
> **biological processes**, conducted in aqueous environments;
> **batteries** using organic electrolytes;
> **geothermal energy** where high temperatures and corrosion are real challenges, with the aggressiveness of the environment being driven by its pH and ionic concentrations.

These applications are now joined by hydrometallurgy, a technique used to enable the recycling of metals from batteries and catalysts.

In parallel, the thermodynamics department is involved in several initiatives on the theme of electrolyte thermodynamics:

- The purpose of the IFP-School chair is to accelerate research with a view to improving the understanding of phenomena and describing them in a macroscopic model. It brings in other IFPEN departments to conduct measurements (phase equilibria, spectroscopy, conductometry).

- Participation in a European ERC project, led by the DTU[a], provides access to high-quality discussions with researchers from several leading universities.

- The EleTher JIP provides a platform for addressing the issue of the transfer of models resulting from research with industrial partners. In fact, a model will only be used on a large scale if it is both available and properly parametrized in a commercial simulator. This partnership therefore involves close collaboration with software sellers.

- Investment in the Prométhée Research Group, bringing together 27 French laboratories, supports IFPEN's new activities in the field of hydrometallurgy.



a- DTU : Danmarks Tekniske Universitet (Technical University of Denmark)

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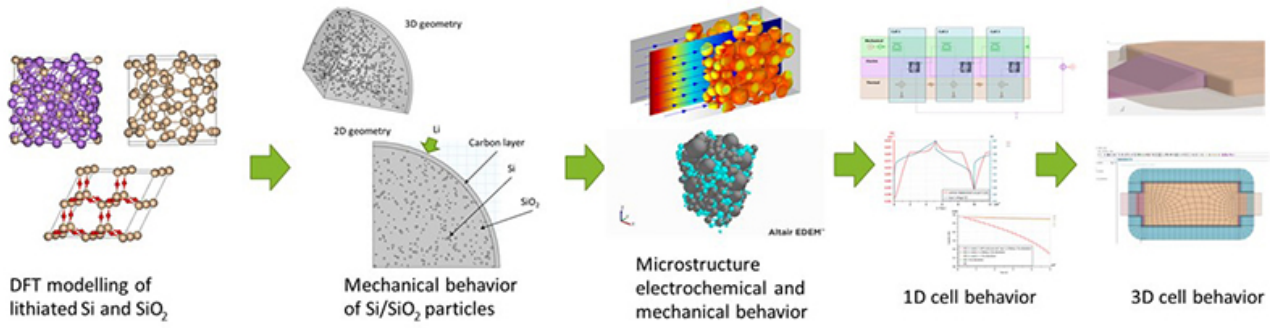
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Electrolyte thermodynamics at IFPEN

For around fifteen years now, IFPEN has been focusing on modeling conventional batteries to represent their nominal operation (electric and thermal behavior during normal operation), throughout their lifetime [1-2] (evolution of performance associated with degradation mechanisms) and in the event of thermal runaway (failure, improper use) [3–5].

Battery technologies, for instance lithium-based, are constantly evolving in order to increase their energy density, reduce their cost and improve their operational safety. At the same time, these multiple evolutions require continual adaptation in terms of the nature[a] and structure[b] of the models used to describe their behavior.

One of the best examples of these evolutions concerns the significant volume variations of electrode materials, during charge and discharge cycles. These lead to variations in internal mechanical stresses and changes in the interfaces between components, with an impact on battery performance. This is the case for high-performance batteries in which the negative electrode contains silicon[c] and for which this mechanical dimension needs to be added to the multi-physical and multi-scale models previously developed [2]. This is the aim of the European MODALIS[d] project (figure), coordinated by IFPEN, and the PSPC-Régions[e] Auranode project, conducted in partnership with Enwires. The new 4th generation of lithium batteries, also known as “all-solid-state” batteries[f], is also concerned by these evolutions. This “all-solid-state” generation will be studied within the framework of the European HELENA[g] project, in which IFPEN is leading modeling activities.



Approach applied within the framework of the MODALIS project

Models can also be enhanced by better consideration of the mechanisms at electrode/electrolyte interfaces in these different systems (liquid/solid or solid/solid interface). The scale of the reactions at these interfaces requires the use of molecular modeling approaches, which is the focus of PhD research conducted in partnership with ENS Lyon engineering school and Stellantis.

Moreover, improving the safety of new battery technologies requires the modeling of phenomena that can cause thermal runaway or system degradation. The model developed in IFPEN describes the exothermal degradation reactions affecting the battery components. It can simulate the cell’s temperature, voltage and internal pressure evolution during thermal runaway. It can also be used to study the propagation of this thermal runaway between the neighboring cells in a battery pack. Lastly, it is coupled with an aging model, so that the impact of aging on battery thermal stability can now be taken into account. [6].

For all these developments carried out by IFPEN, a multi-physical and multi-scale approach is used based on combining electrochemistry with diffusion, interfacial, thermal and mechanical phenomena. This approach is already used to study other energy storage systems, such as flow batteries, or fuel cells [7].

a- Electrical, electrochemical, mechanical, chemical
b- Coupled/uncoupled, complex/simplified
c- Generation 3b (high capacity/high voltage)
d- MODelling of Advanced LI Storage Systems
e- Strategic research and development project for competitiveness
f- In which lithium is used for the negative electrode and where the liquid electrolyte is substituted with a solid electrolyte
g-Halide solid state batteries for electric vehicles and aircrafts

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Battery behavior: a complexity best taken into account by modeling

Anchor lines, the majority of which are carbon steel cables, are essential components for the stability of offshore floating structures, such as those supporting wind turbines. To overcome the risk of breakage during service, redundant lines are generally incorporated at the design stage, which adds significantly to the cost. The challenge is to improve cable reliability in order to reduce the need for redundant lines, via better prediction of their fatigue life.

Steel cable fatigue is a multi-scale phenomenon. On a large scale, it depends on tension and bending variations associated with stresses exerted by the wind, waves and currents. On a metric scale, behavior depends on friction between the wires making up the cables (Figure 1). Finally, on the millimetric scale of contact between wires, the phenomenon depends on the local mechanical load and the environment (air, seawater, grease).

The approach adopted by IFPEN, in partnership with the LMPS laboratory[a] at the Ecole Normale Supérieure and the LTDS laboratory[b] at the *Ecole Centrale Lyon* engineering school, consisted in replacing the existing empirical prediction of fatigue resistance with multi-scale modeling based on physics. On a macroscopic scale, the critical zones of the anchor line are determined with aero-servo-hydro-elastic simulation[c] of the floating structure, which takes into account wind, waves and ocean currents. With variable tension and bending, a load is deduced from a mesoscopic FEM[d] model (around ten meters) of the cable, which predicts slip and stresses between the wires (Figure 2). On the scale of contact between the wires, modeling makes it possible to verify compliance with a fretting-fatigue[e] criterion identified experimentally taking into account the environment (Figure 3) and which corresponds to the partial slip/total slip transition[f] .

The research conducted has led to significant advances for the simulation of the behavior of steel anchor lines and a better understanding of the fretting-fatigue behavior of the wires contained within them.

For the first aspect, on a mesoscopic scale, the use of the new FEM model generates considerable savings in terms of calculation times. Research currently underway is targeting further decreases through reduced-order modeling via the PGD-LATIN [3] method implemented by LMPS.

For the second aspect, in addition to highlighting the existence of a critical slip, fretting tests demonstrated the decisive yet complex and sometimes counter-intuitive influence of the surrounding environment. Grease has a positive but limited effect, because its high viscosity limits the penetration into the contact. Seawater has two opposite effects : in one hand it reduces damage by its lubricant properties, in the other hand, it increases it by its corrosive action. Depending on the loading, one or the other mecanism is preponderant (figure 4). The tests in seawater were done in an aerated cell, with control of the electrochemical potential.

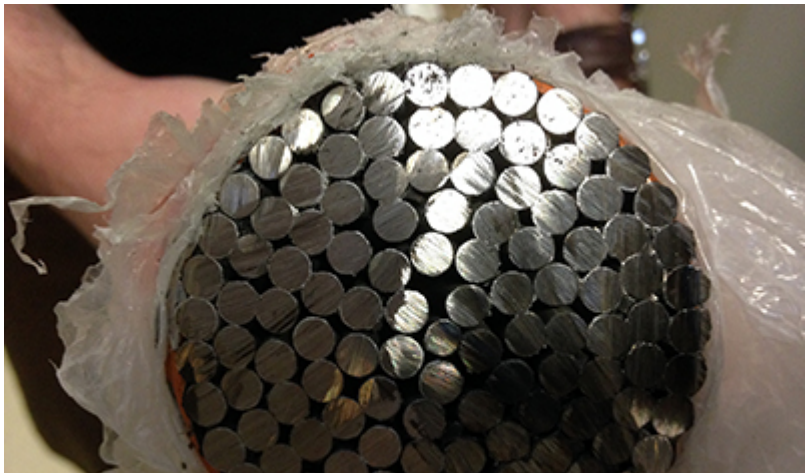


Figure 1: Section of a steel anchor cable

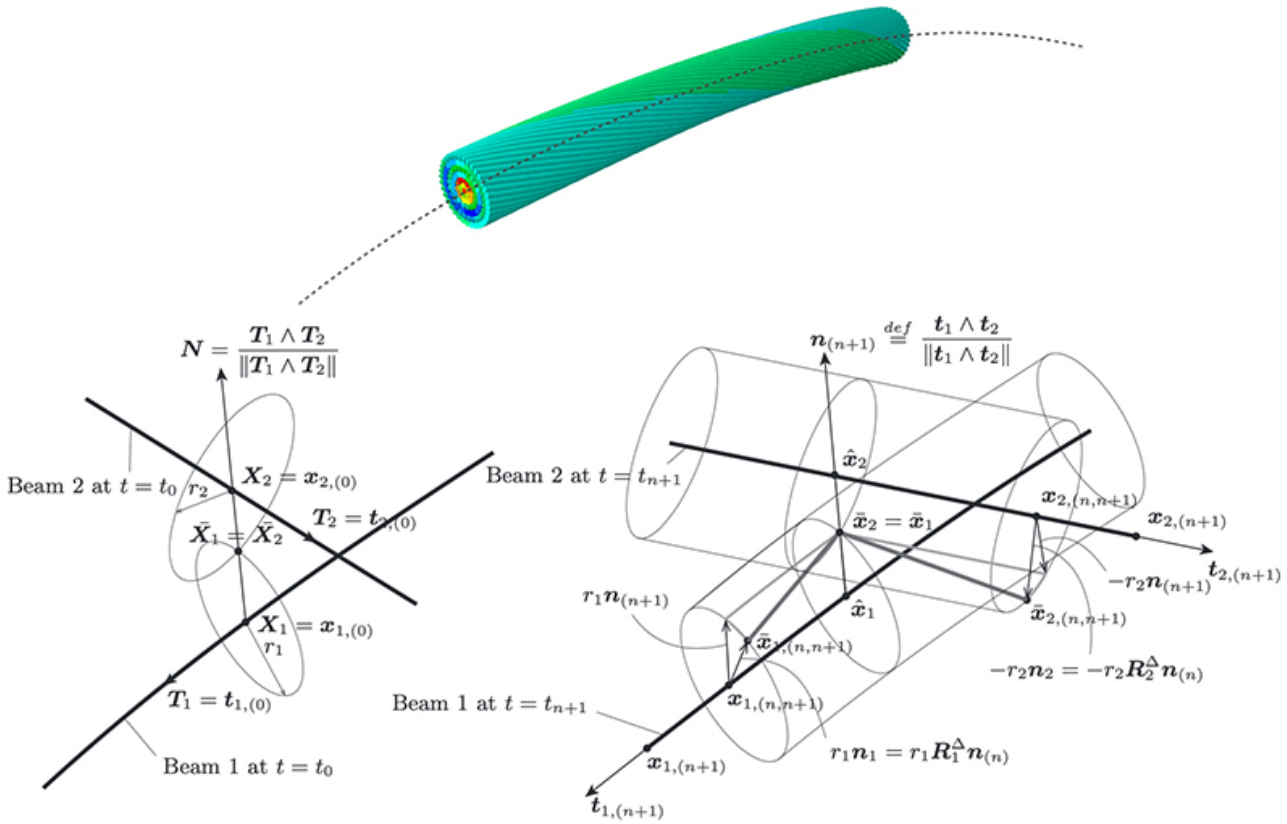


Figure 2: Finite element model of a cable with contact and friction [1].

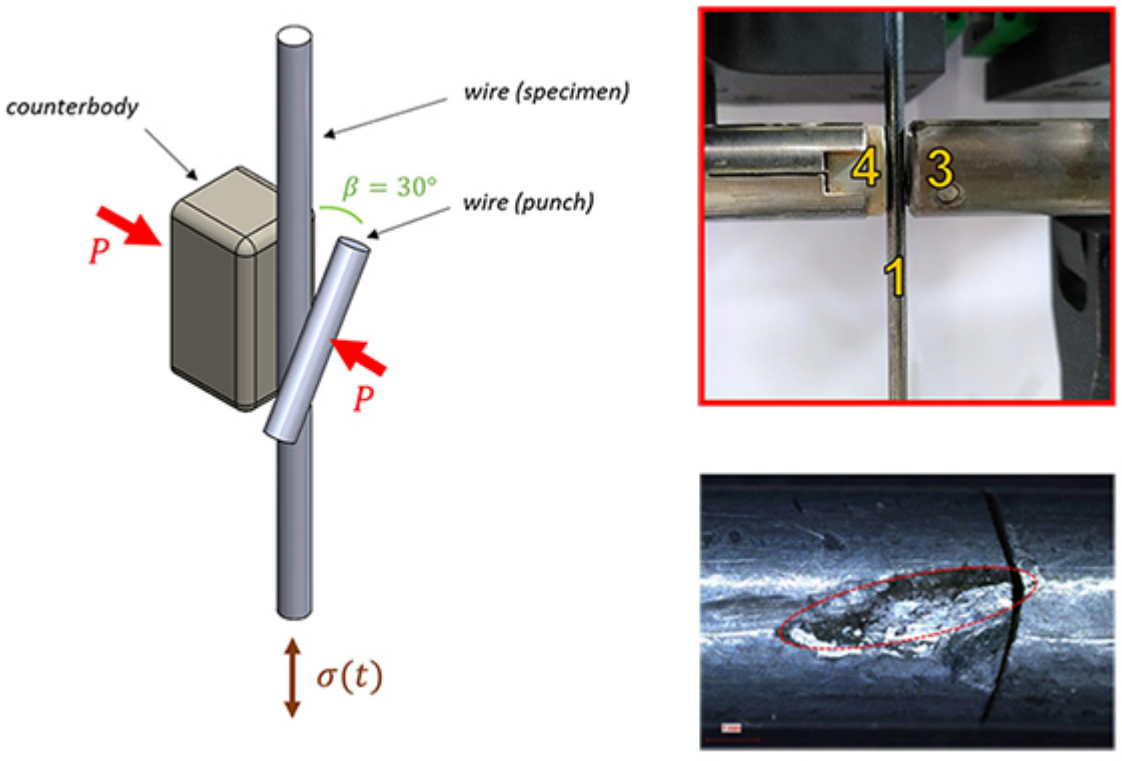


Figure 3: Fretting-fatigue test on a cable wire at the LTDS [2]

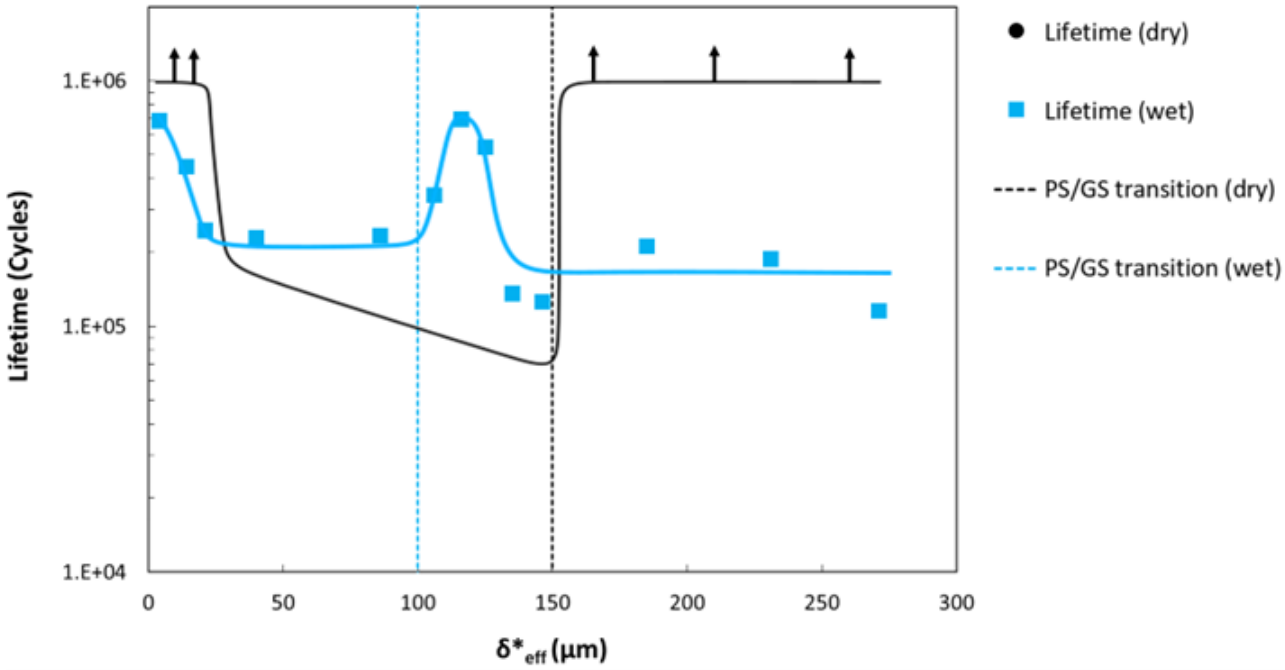


Figure 4: Fretting-fatigue curves (sliding on x-axis, cycles to failure on y-axis) for steel-steel contact dry (black) and in seawater (blue). PS/GS : « partial slip / gross slip »

a- Laboratoire de Mécanique Paris-Saclay (Paris-Saclay Mechanics Laboratory)
b- Laboratoire de Tribologie et Dynamique des Systèmes (Tribology and System Dynamics Laboratory)
c- Aerodynamic + servo for the controller + hydrodynamic for the immersed elements + elastic for the structure.
d- Finite element model: beam-type in contact with friction in finite rotations and small displacements.
e- Fretting fatigue is the phenomenon on crack initiation and propagation
f- Partial slip: part of the contact surface slips, the other remains stuck; total slip: the entire contact surface slips

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“From material to structure” modeling: the case of anchor cables for offshore wind, in corrosive environment

Wind energy accounts for an increasing share of the energy mix thank to the construction of wind farms designed to reduce investment and operating costs. However, within a farm, downstream wind turbines are exposed to the wakes of upstream rotors, leading to a reduction in their production and an increase in their fatigue stresses. Moreover, wind turbines operate in a complex environment, known as the atmospheric boundary layer (ABL), which interacts with these wakes. This zone of the atmosphere is subjected to large-scale effects (pressure gradients, Coriolis effects[a]) as well as local effects, such as thermal exchange with the ground and the surrounding topography.

For numerical simulation (as illustrated in Figure 1), a real challenge exists, associated with the multi-scale nature of the problem: it is necessary to analyze and model the impact of kilometric phenomena (atmospheric turbulence) on wakes, the origin of which is on a metric scale (flow around a blade).

To address this problem, a research partnership was set up with the CNRM (French National Meteorology Research Center) hinged around the Meso-NH simulation tool. The CNRM integrated precise ABL modeling into the tool, including the various phenomena of importance for wind energy technology, such as turbulence, thermal stratification and topography[1]. For its part, IFPEN implemented representative wind turbine models in the tool, based on actuator line and disc approaches [2] [3].
As a result of these changes, researchers were able to use Meso-NH to study wake behavior [4][5][6] and develop analytical models that have since been incorporated in FarmShadow^{TM}, the wind farm design tool developed by IFPEN.

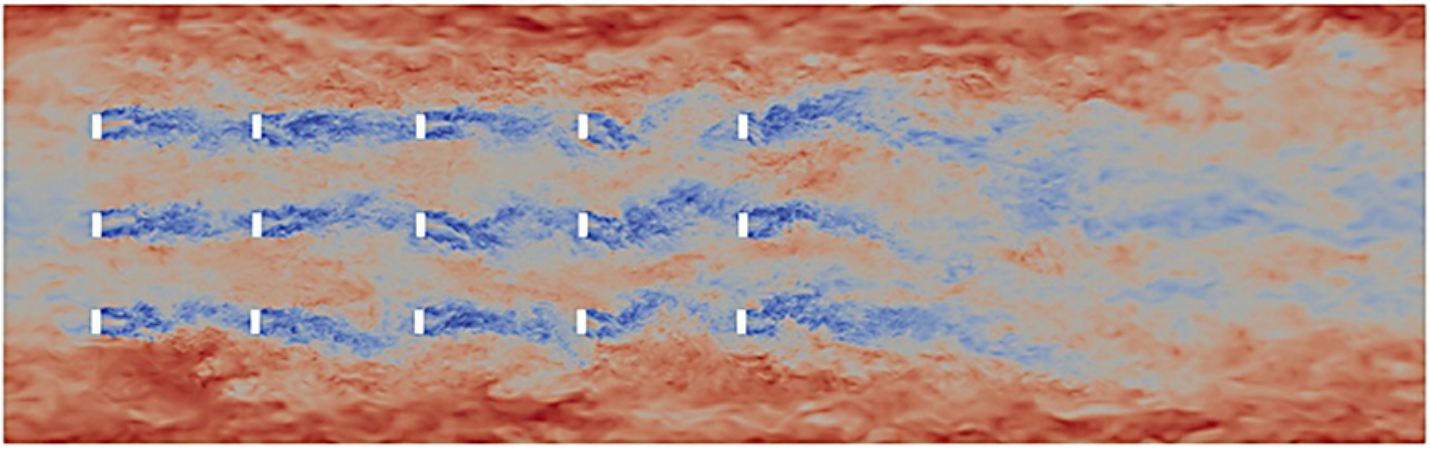


Figure 1: Horizontal cross-section of the wind field, simulated by waLBerla[b] [IFPEN/FAU-Erlangen[c]], crossing a wind farm

The resulting improvement compared to existing simulation tools is that FarmShadow^{TM} makes it possible, in a few seconds of calculation time, to estimate the production of wind farm. Combined with IFPEN's optimization methods, it also makes it possible to maximize a farm's overall production by adjusting wind turbine placement.

Moreover, new analytical models of unsteady wakes interacting with the ABL will be employed in the DeepLines Wind^{TM} aero-hydro-servo-elastic computational code, used to design a wind turbine within

a farm.

Regarding high-fidelity simulations, one sticking point still to be overcome is that of calculation time: conducted by LES[d], simulations arelimited from a practical point of view to a few configurations including only a few turbines. An alternative approach, based on Lattice Boltzmann methods (LBM), may make it possible to overcome this challenge and is currently the focus of research (figures 1 and 2), in partnership with Erlangen University [7]. By exploiting the capacities of computer graphics cards, early results show a 400-fold reduction in calculation time. Work is in progress to implement in this solver the physical models necessary for the simulation of CLA.

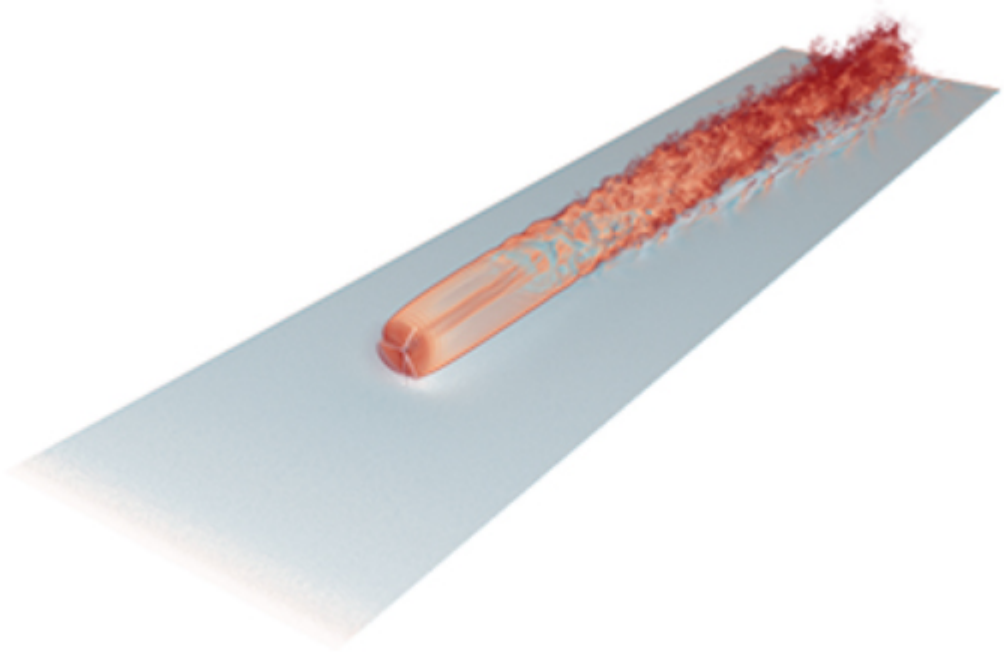


Figure 2: LBM simulation of a horizontal wind turbine
(IFPEN/FAU-Erlangen)

a- Coriolis force: an inertial force that acts perpendicular to the direction of movement of objects in motion in a medium itself rotating uniformly
b- waLBerla: Widely applicable Lattice Boltzmann from Erlangen: massively parallel framework for multi-physical applications
c- FAU-Erlangen: Friedrich-Alexander-Universität Erlangen-Nürnberg

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