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ANNEX 1: RESEARCH PLAN

PERSONAL INFORMATION

First Name:	*Giuseppe Antonio*
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PROJECT INFORMATION

Project Acronym:	*HM^2F*
Title of Research Project:	*New design for hierarchical multifunctional membranes: theoretical and numerical characterization.*
Research Focus Area:	*HPC*

Host University:	*EPFL*
Host University Department:	*STI-IGM-LFMI*
Supervisor:	*François Gallaire*

Co-Host University:	*DTU*
Co-Host University Department:	*Department of Physics*
Co-Supervisor:	*Tomas Bohr*

1. ABSTRACT

The state of the art of filtering and membrane processes evidences the existence of limitations, at theoretical and practical levels, related to their efficient performance. Techniques to analyze the behavior of fluids flowing through membranes and the performances of the membrane itself, are essentially based on microscopic models with experimental justifications, or on ad hoc models derived by merging different theories (cf., for instance, Ledesma-Duran et al., 2017). The purpose of this project is the design of multi-scale hierarchical membranes, characterized by the concept of modularity, being nowadays one of the fundamental needs in technological adaptation (cf. Schilling, 2000), and of the development of a self-contained theoretical and numerical framework to analyze them, based on homogenization theory. The High Performance Computing (HPC) aspect, treated here in its multi-physics variant, is fundamental to the success of the project. Resources-consuming direct numerical simulations of fluid flows and particle transportation across the studied membranes will be calculated, to validate the multi-scale model developed. As final result, an easy-to-use and computationally light reduced order model to analyze the dynamics of fluid solvent and solutes through membranes will be delivered to the scientific community. This project represents the opportunity for the project investigator (PI) to work in a top level institution where he can extend the range of application of multi-scale modelling, facing theoretical problems such as homogenization through interfaces, and a chance to acquire important expertise in multi-scale techniques, responding to its purpose of analyzing physical world starting from first principles by upscaling from a molecular to a universal level.

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2. PROJECT DESCRIPTION

2.1 State of general and own research;

Membranes used for filtering processes exhibit different characteristics, according to their functions (e.g. desalination, recovery of precious metals, sterile filtration, food processing, paint and solvent recovery, petroleum refining and other industrial, medical, and environmental applications, cf. Mohanty & Purkait, 2011; for a review). The filtering properties depend, among other properties, on the dimensions of the pores, and they range from particle filtration (able to filter human air, fat micelles and activate carbons, with typical characteristic pore size between $10\mu m$ to $1mm$) to osmosis (essentially used in desalination, with typical pore size of about $1nm$), passing throughout microfiltration, ultrafiltration and nanofiltration. In nature, plants depend heavily on osmosis and filtration for their water and sugar translocation.

In a recent paper, Park *et al.* (2017), highlighted the need of a better understanding of the mechanisms behind these processes, responsible of the trade-off between permeability and selectivity of membranes, in order to exceed the existing bounds. One of the critical points in membrane design is to combine mechanical resistance to stresses with a determined permeability and selectivity. When particles encounter a membrane, if their mass is too high, a rupture of the membrane may happen. Moreover, in pressure driven backwashing for cleaning purposes (either in micro-, ultra- and nano-filtration, cf., for instance, Ramon *et al.* 2010; Rho *et al.* 2013), high pressure gradients act

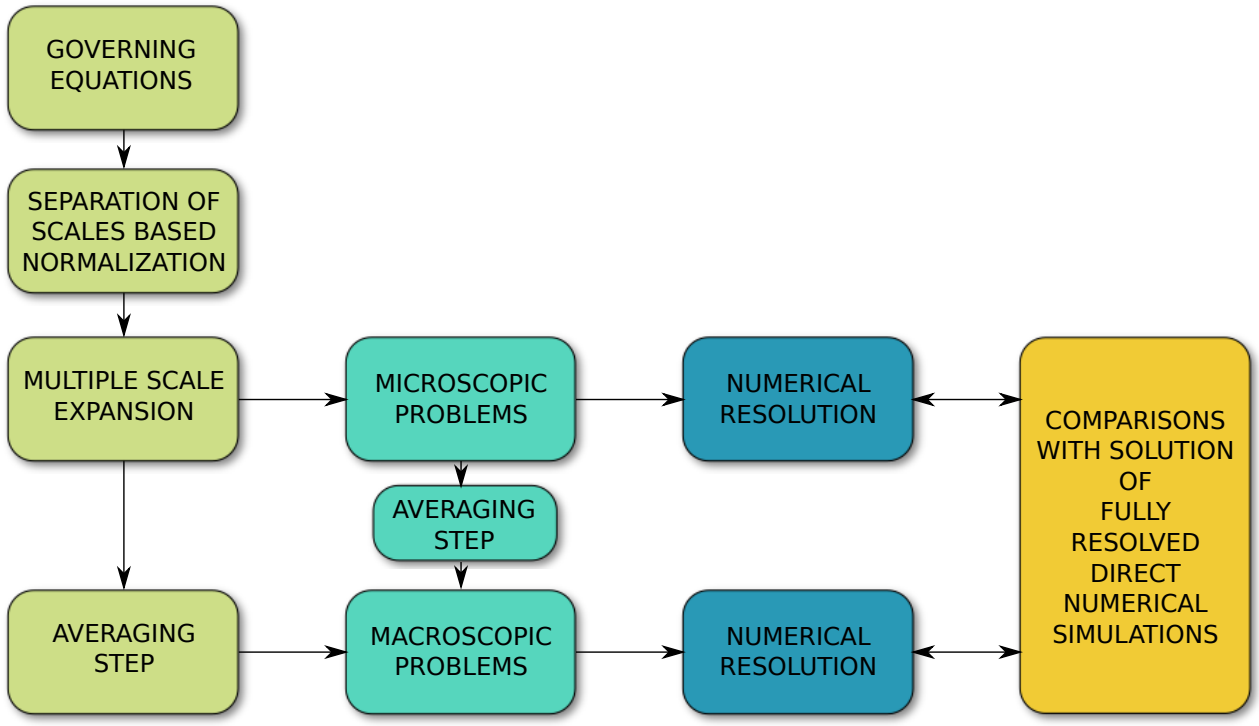

Figure 1: Workflow of the homogenization technique used in Zampogna & Bottaro (2016,2017) , Lacis et al. (2017), Zampogna et al. (2018) to deduce macroscopic models from a microscopic description of a physical phenomenon. In these works theoretical models found applying homogenization to first principles (called governing equations in the figure) are accompanied with their numerical solution and validation against direct numerical simultations (DNS). The resulting model, composed by microscopic problems, to be solved over a microscopic cell representative of the microscopic porous, poroelastic or rough structure, and by macroscopic equations, valid over homogeneous domains without distinctions of phase, are computationally light with respect to DNS.

on the membrane, causing a possible breach. Many works with the purpose of reinforcing membranes via the use of layers of crossed fibers have been proposed, most of them presenting only experimental results, as in Chou *et al.* (2013) et She *et al.* (2016).

A systematic analysis of the wide literature on membrane technologies shows that experimental works are more common than theoretical and numerical studies and the molecular, chemical, biological point of view is preferred to continuum fluid mechanics (cf. for instance, Farahbakhsh *et. al.* 2017). Studies aiming to describe the behavior of fluid flows through membranes from a full continuum point of view, are generally based on early works of Kedem and collaborators, such as Kedem & Katchalsky (1958) and Spiegler & Kedem (1966). In these works, the equations describing the fluid flows through membranes resemble Darcy's law, with an added pressure gradient which depends on different distributions of the solute across the membrane. In these equations, ad hoc tuned parameters appear, such as the permeability associated to the fluid and to the solute, often deduced with the use of experimental laws. Among the works that use these approaches Bacchin (2017) can be cited. Cardoso & Cartwright (2014) tried to include in the same continuum model a microscopic characterization of the solvent-solute behavior via the introduction of a parameter found by solving problems at molecular scale. A further step has been done by Ledesma-Duran *et al.* (2016, 2017): although they assume the validity of a set of continuum equations taken from different theories, they find a strategy to characterize the intrinsic parameters related to the triple solvent-solute-membrane by solving pore-scale problems for some specific geometries.

In conclusion, the lack of a systematic and methodical analysis of continuum fluid mechanics and solutes' diffusion across membranes, in a theoretically unified sense, can be evidenced from a review of the state of the art (cf. Ho & Sirkar, 1992). In order to address the critical issues described at the beginning of the section, the present project tries to compensate this lack by merging a self-sufficient theoretical model and its numerical solution, accompanied by a validation, against direct numerical simulations (DNS), able to describe the whole system solvent-solute at the smallest scale.

The essential tools necessary to fulfill this objective are already present in some of the works of the PI, which essentially used multi-scale homogenization (whose procedure is sketched and briefly described in figure 1) to model fluids flowing through rigid and deformable porous media, or above rough surfaces. In these studies, a macroscopic description of the physical phenomenon is provided, containing some characterizing tensors, such as the permeability, the effective elasticity and porosity, or the slip tensor, which solve problems over sample microscopic domain of the size of the pores. The resulting reduced order model, completely autonomous (i.e. there is no need to resort to experimental data or additional theories in order to close the problem), is always corroborated with its numerical solution and compared with DNS of the complete multi-scale phenomenon for validation purposes. In general these models, being composed by averaged equations, are computationally lighter than DNS since they are valid over homogeneous domains, where there is no need to simulate different phases. To asses a direct link between these works and the physical phenomenon analyzed in the present project, a few things must be noticed:

- Homogenization is widely utilized to study rigid and deformable porous media.
- A homogenization technique has been modified in Zampogna et al. (2018) to be suitable to analyze fluid flows over thin rough layers.
- Membranes can be seen as rigid or deformable porous media, very thin along the flow direction.

As consequence of these aspects, a framework to study fluid flows and solute transport across membranes is developed as described in the next section, in order to satisfy the urgent needs which are not treated in the present literature.

2.2 Detailed research plan including aims; questions to be answered; hypotheses; methods and risk management;

In section *2.1* the need to design and model membranes in order to quantify and enhance some of their properties was highlighted. The present section proposes a strategy to examine in depth the critical issues noticed before, by developing a theoretical and numerical framework to simulate the solvent's and solute's flows across membranes. Then, an original new membrane will be proposed in answer to the needs mentioned in section 2.1, with relevant implications in industry, as explained in section 2.4. To these purposes, the most relevant properties of the developed model must be:

- Generality; a description of the phenomenon for every kind of solutes, solvents and membrane's structures and material is needed. Hence, in the hypotheses, as it will be explained later, only first principles defining the nature of matter are assumed.
- Applicability and concreteness; the theory must be able to analyze practical cases, but, even more, it must give new insights in membrane technology (later on this point will be clearly addressed by a membrane design partially defined on the basis of the theory itself).
- Simplicity, ease of use; since a useful tool must be delivered to the scientific community, its salient ingredient is the easiness in the formal comprehension, from a theoretical mathematical point of view, and the easiness of numerical implementation. To this scope a reduced order model, able to run on every machine without computational limitations, is proposed.
- Multi-physics; a complete description of the physical reality cannot dispense with this aspect.

In particular, the attention must be focused on the last two aspects which, nowadays, are still opposite, being the technical implementation of the coupled fluid-solid behavior which is computationally consuming and hard to implement.

In order to methodically address these aspects and rationalize the feasibility of the project, the project is divided in two large parts (carried out at EPFL or DTU on the basis of the tasks defined in section 2.3 and the host's and co-host's skills mentioned in section 2.5) These parts are consecutive in time. Since the main goal consists on the design and analysis of hierarchical multifunctional membranes, the first step is to carry out an as "universal" as possible model to analyze generic membrane phenomena. A *simple* membrane will be analyzed with the homogenization technique used by the PI in the works cited in section *2.1*. Here, the term *simple* stands for the membrane sketched in figure 2, i.e.:

- A macroscopic length scale can be associated to the physical phenomenon (D, in figure 2).

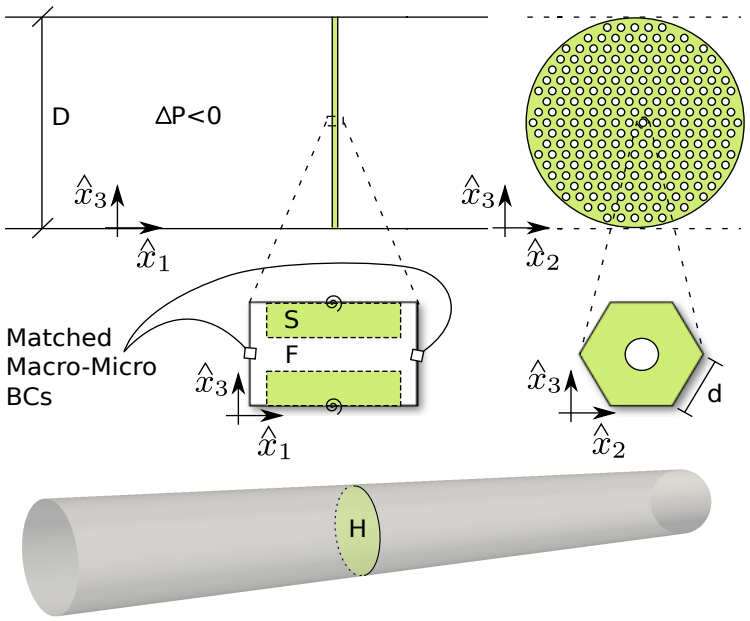

Figure 2: Macroscopic (top and bottom) and microscopic (center) geometrical description of a simple membrane. The boundary conditions imposed at the small scale problem are indicated.

- A microscopic length scale can be associated to the physical phenomenon (d, in figure 2).
- The pores can have a generic shape, but they must be placed with a periodic arrangement in the cross flow plane (a hexagonal arrangement of cylindrical pores is adopted in figure 2).
- Chemical reactions are not considered in the model, only fluid mechanics is simulated. However, since no restrictions about macro- and microscopic boundary conditions are present, sources or sinks for the solutes can be simulated.
- Linear elasticity of the membrane is admitted, allowing to evaluate its stiffness.

To explain the procedure used in the present analysis, the configuration described in figure 2 is considered. A cylindrical channel, filled with an incompressible fluid, satisfying the Navier-Stokes equations (NSE), is separated by a *simple* membrane. The convection-diffusion equation governs the distribution of the solute while the balance equations of a linear elastic solid are used to quantify the stresses acting on the membrane. As already mentioned in the previous section the final scope of this step is to simulate the presence of the membrane as a point-wise constraint along $\hat{x}_1$. From a three-dimensional point of view this is equivalent to deducing a modified governing equation valid over a cross sectional plane intersecting the channel, the H region in the bottom of figure 2. With these hypotheses the procedure sketched in figure 1 can be applied. First of all, a microscopic elementary cell, also called representative elementary volume (REV), must be selected (cf. the second row of figure 2, where the REV is represented by projections over two principal orthogonal planes). The behavior of the unknown fields in this cell, due to the periodic arrangement, is representative of the flow behavior in the other cells composing the membrane. After a proper normalization of the point-wise equations, based on the small parameter ε, in this case defined as $\varepsilon = \frac{d}{D}$, a multiple scale expansion of the equations is performed. This essentially means that each unknown field f_i (e.g. fluid velocity and pressure, solute distribution and displacement of the membrane) can be decomposed into an series of increasing powers of ε times some other functions, which depend on a fast spatial variable $\boldsymbol{x}$ (which scales with d) and a slow spatial variable $\boldsymbol{X}$ (which scales with D):

$$f_i(\boldsymbol{x},\boldsymbol{X}) = \sum_{n=0}^{\infty} \varepsilon^n f_i^{(n)}(\boldsymbol{x},\boldsymbol{X}).$$

Physically speaking this is equivalent to saying that, due to a strong separation of scales, i.e. $d \ll D$, the phenomenon can be decomposed into a microscopic part, where variation over d can be noted and a macroscopic part, where only variation over D can be appreciated. The membrane is responsible of the variations over d which otherwise should not be present under certain flow's conditions and solute's properties. After this step, the new unknowns are $f_i^{(n)}(\boldsymbol{x},\boldsymbol{X})$, instead of $f_i(\boldsymbol{x},\boldsymbol{X})$. PI has shown in Zampogna & Bottaro (2016,2017), Lacis *et al*. (2017) and Zampogna *et al*. (2018) that the so called *leading order approximation* $f_i^{(0)}(\boldsymbol{x},\boldsymbol{X})$ already furnishes a good estimation of the original unknowns $f_i(\boldsymbol{x},\boldsymbol{X})$. In force of this, only the leading order equations are solved over the REV, with periodic boundary conditions along the tangential to the membrane directions. Attention must be paid to the conditions imposed over the boundaries, denoted in figure 2 as "matched macro-micro bcs". They represent the macroscopic forcing on the microscopic problem or, mathematically speaking, a basis for the space of the solutions of the microscopic partial differential equations. If $B^{(j)}$ is the j-th forcing, the leading order approximation of each microscopic unknown field $f_i^{(0)},\ i = 1,\dots,M$ can be written as

$$f_i^{(0)}(\boldsymbol{x},\boldsymbol{X}) = \sum_{j=1}^{J} a_i^{(j)}(\boldsymbol{x}) B^{(j)}(\boldsymbol{x},\boldsymbol{X}). \qquad (1)$$

The solution of the microscopic problems can be determined finding $a_i^{(j)}$ which are the coefficient of $f_i^{(0)}$ with respect to the basis $B^{(j)}$. These coefficients solves the microscopic equations on the REV, found by substituting the new form of $f_i^{(0)}$ in the original microscopic problems. In the present case,

the forcing terms derive from conservation of mass and momentum (or exchange of them if sources or sinks are allowed). Finally, the macroscopic model is found by considering the spatial average of equation (1), defined as the integral over the REV with respect the variable $\boldsymbol{x}$. After averaging, each cell collapses in a macroscopic point $\boldsymbol{X}$. For this reason the final model is a one point constraint along $\hat{x}_1$, valid on the circle H where there is no more distinction between the solid and the fluid phase. It will reach its maturity via validations by means of comparisons with the previous literature (cf. for instance Kedem & Katchalsky, 1958 and Mikulecky, 1972 for a comparison with previous theoretical model or Gruber et al. 2011 for a comparison with previous numerical results) and of comparisons with respect to DNS, resolved at the smallest scale present in the physics of the phenomenon, carried out with the HPC resources available at the host or co-host institution (e.g. FIDIS at EPFL or Niflheim at DTU). Finally, a macroscopic tool to analyze fluid flows through membranes is delivered to the scientific community, where some microscopic tensors are presented, able to characterize the microscopic behavior of the fluid flow and solute transport for every kind of pore shapes and membrane material. This model is valid over a homogeneous domain, where there is no distinction between the solid and the fluid phase.

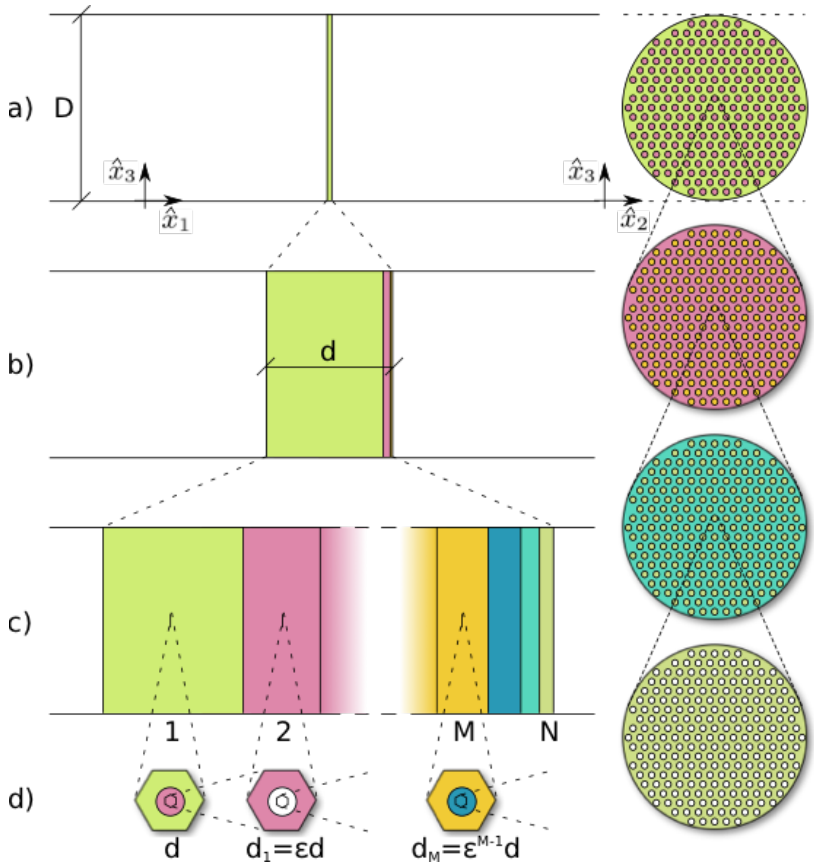

Figure 3: Frame a): stratified recursive membrane in a cylindrical channel represented on the orthogonal planes $(\hat{x}_1, \hat{x}_3)$ and $(\hat{x}_2, \hat{x}_3)$. Frames b) and c): zooms on the membrane along $\hat{x}_1$. At a linear scale the small-scale layers composing the membrane cannot be seen and a conceptual logarithm scale must be used along $\hat{x}_1$ (frame c). In this representation the aspect ratio between each consecutive layer can be appreciated and the nested fractal-like structure of the membrane is highlighted (frame d and right column of the figure). The particular design of the membrane requires a recursive homogenization technique to be analyzed. Homogenization can be applied to the governing equations of the flow N times, resulting in a macroscopic model simulating the presence of the membrane, which acts on the fluid flow as a pointwise constraint along $\hat{x}_1$.

Beyond this first step, which theoretically represents the breakout of the limitations of application of homogenization in bounded domains, the design and analysis of *multifunctional* membranes can be carried out with a little effort. The term *multifunctional* is antithetic to the term *simple*, previously introduced, being characterized by the following factors:

- A macroscopic length scale can be associated to the physical phenomenon (D, in figure 3).
- N microscopic length scales can be associated to the physical phenomenon (d_n, in figure 3); the relation $d_{n+1} = \varepsilon\, d_n$ between each scale is valid; each of them ideally corresponds to a function of the membrane (from the micro-scale for micro-filtration to the nano-scale for nano-filtration passing through the ultra-scale for ultra-filtration, for instance).
- Chemical reactions are not considered in the model, only fluid mechanics is simulated. However, since no restrictions about macro- and microscopic boundary conditions are present, sources or sinks of solutes can be simulated.
- Linear elasticity of the membrane is admitted, allowing to evaluate its stiffness.

As in case of *simple* membranes, the starting point consists of the continuum NSE, the convection-diffusion equation for the solute distribution and the balance equations of a linear elastic solid de-

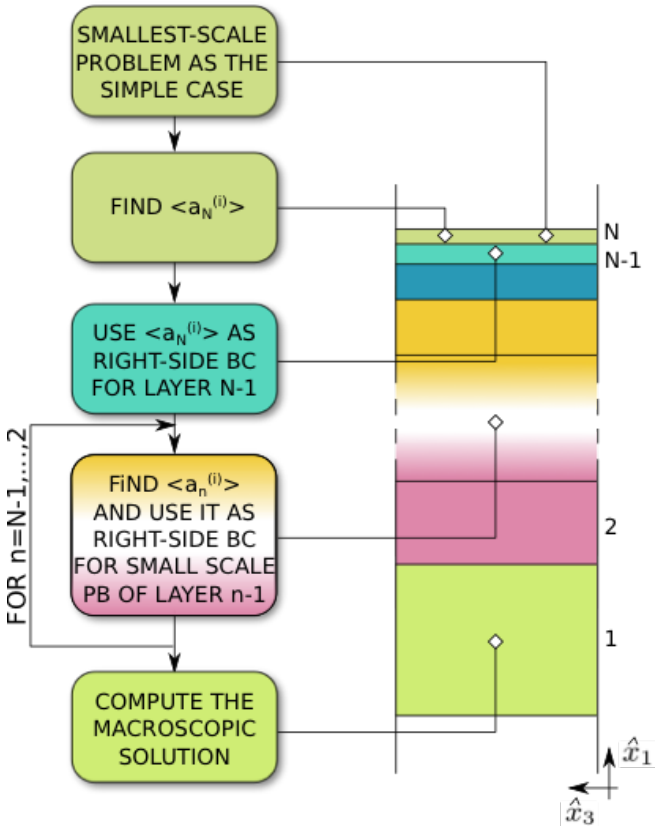

Figure 4: Workflow diagram of the recursive procedure used to analyze multifunctional membranes.

scribing the displacement of the membrane. A procedure based on that described in the previous case is implemented: the previous process must be intended as a *basic module* which is recursively repeated. The differences in the procedure are due by the geometry of the *multifunctional* membrane, built as a modular overlay of N completely identical layers, each of them with thickness and pore size rescaled of a factor ε with respect to the left-adjacent one, as described in figure 3. To avoid confusion, since there are several scales, the length D is called macroscale, d_N is the smallest scale and each length d_n with $n = 1, \dots, N-1$, is an intermediate scale. The intermediate scale d_n has an upper-scale d_{n-1} (on the left in figure 3) and a lower scale d_{n+1} (on the right in figure 3). For

fixed n, the upper scale represents the macroscale for the n-th problem. A successful procedure will allow to exchange informations between the scales. To obtain a macroscopic model the analysis is started from the smallest scale layer. Since by construction $d_N = \varepsilon\, d_{N-1}$, the homogenization procedure explained for *simple* membranes can be applied between level $N-1$ and N. Anyway, no upper-scale description of the N-th layer of the membrane is required. Only the microscopic problems for the coefficients $a_{i,N}^{(j)}$, $j = 1, \dots, M$, N standing for the N-th layer and i representing the i-th unknown field, must be solved. Their spatial average, denoted with $< a_{i,N}^{(j)} >$, is then used as right-side boundary condition for the microscopic problem at the upper-scale $N-1$, i.e. $a_{i,N-1}^{(j)} =< a_{i,N}^{(j)} >$. Since also for layer $N-1$ the relation $d_{N-1} = \varepsilon\, d_{N-2}$ is satisfied by construction, the same left-side boundary condition of the microscopic problem in case of *simple* membrane can be utilized. Neither for this level the upper-scale model must be deduced, but only $< a_{i,N-1}^{(j)} >$ must be computed to furnish right-side boundary conditions for the next problem. This procedure is iterated $N-2$ times up to reach level 1. At this point we solve for the last time the microscopic problems with modified right-side boundary conditions and then we take the average of equation (1) to write the macroscopic model. The workflow of the whole procedure detailed here is described in figure 4. The particular original structure of the membrane does not make possible comparisons with respect to the previous literature and the validation of the model via the use of DNS becomes fundamental. In this case their computational complexity increases by increasing the number of the layers N and huge HPC resources are needed to carry out the results. Once the model is validated it will allow to analyze also similar simplified configurations present in literature, where nanoscopic membranes are reinforced by layers of crossed fibers and comparisons with respect to the stiffness of the developed membrane will be done, giving guidelines in the improvements of the capabilities and performances of these systems.

Risk assessment: A critical point of the procedure, in *simple* and *multifunctional* membranes, consists in transferring the macroscopic quantities as boundary conditions for the microscopic problem. This procedure, however, has been successfully applied by the PI in Zampogna *et al.* (2018) to analyze incompressible fluid flows over micro-rough surfaces and the only difference with respect to the present case is that, here, a penetration of the surface is allowed. Another critical point is linked to the semi-permeability of membranes. In processes at the nanoscale, like osmosis, the pores are sufficiently small in order to block determined molecules with a certain size. The strategy proposed here, even if it allows to determine an effective diffusion coefficient for the solute, which accounts for the obstruction due to the solid structure, will be probably not able to take into account a full blocking of certain solutes, as discussed in Dölger *et al.* (2014), unless artificial expedients such as ad hoc sources or sinks or filtering functions for the diffusivity (used, for instance, by Kedem & Katchalsky, 1958) are introduced. Since the use of these expedients is not in the philosophy of the project, at a certain point the PI will evaluate if it is necessary to go up to the molecular scale to develop, via an upscaling technique similar to homogenization, continuum equations starting from the Boltzmann equations, describing the behavior of the solvent-solute at a molecular level, to deduce continuum properties to be inserted in the macro-model in order to be able to simulate the block of certain chemical species.

2.3 Schedule and milestones;

In this section the scheduling of the main tasks needed to carry out results in an efficient way is synthetically exposed. The following tasks have been identified:

- TASK 1: Theoretical modelling of the physical phenomenon. This includes also a precise definition of the physical starting hypotheses, possible only after an in-depth study of the

literature in the topic, the definition of the boundary conditions to transfer macroscopic information to microscopic problems, the application of homogenization to *simple* membranes.

- TASK 2: Numerical resolution of the microscopic equations. This includes the implementation using OpenFOAM, the selection of some fundamental parameters to be varied in the study, the resolution of the equations, the validation with the previous literature. Numerical resolution of the macroscopic equations, implemented using OpenFOAM (or MatLab/Python for some simplified case), the resolution of the equations and the validation by comparing with the previous literature, where present. DNS of the same macroscopic configurations to validate the model are carried out with OpenFOAM in this task.
- TASK 3: Analysis of the results. This includes also the application of the model to cases present in literature in order to proceed with the identification of the aspects to be changed in the physical configuration to enhance determined properties of the membrane, followed by an optimization with respect to the parameters identified.
- TASK 4,5 and 6 are the counterpart of Task 1,2 and 3 for the case of *multifunctional* membranes.
- TASK 7: Deliver the produced results.
- TASK 8: Future development and career plan.

Table 1 shows how these tasks are distributed during a 24-months-long postdoctoral fellowship. The gray bands represent some important milestones: once the theories associated to simple and multifunctional membranes are developed and the numerical results carried out, at least two scientific papers in peer reviewed journals will be written. Moreover, task 4 and 8 will give rise to two more papers addressing the practical aspects for which the project is written, i.e. giving criteria and insights to improve the performances of the physical phenomenon and answer to the needs of the state of the art.

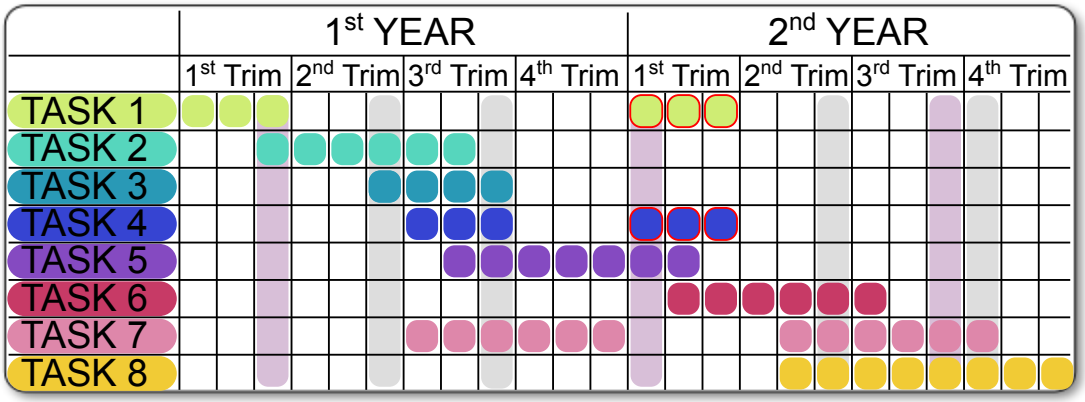

Table 1: Scheduling of the project. The green and blue squares circled in red correspond to eventual further developments of the theory in order to face to the risks described in the previous section. The gray bands correspond to the milestones previously described. The lilac bands represent possible periods spent in the co-host institution to develop or improve the theoretical model and to finalize the project.

2.4 Relevance of the project and expected impacts;

In section 2.2 a hierarchical multifunctional membrane has been designed and analyzed. It satisfies three fundamental criteria: high permeability, high selectivity and strong resistance to external stresses. The high permeability is given by the fact that the thickness of each layer is equal to the pore size, being the pressure loss across the membrane minimized. In the meanwhile, since at least from a potential point of view the number of layers can be increased as you like, without significantly

increasing the thickness of the membrane, also the selectivity can be increased. Being the most selective layer stitched to the others, a high selective membrane has the same effective resistance to stresses as the first low selective layer of thickness d. Once these characteristics, which perfectly responds to the needs of the actual membrane advances, according to Park *et al.* (2017), will be checked thanks to the macroscopic model, this kind of membranes will be delivered to the scientific community giving rise to a class of hierarchical objects which can fit any application. The production of these membranes should not be difficult, neither for the higher layers, due to the diffuse trend of downsizing systems; recent progresses in carbon nano-lattices could be utilized to this purpose (cf. Bauer *et al.*, 2016). Much work should be done by chemical engineers to understand and create the best stitching conditions for each layer. If in this process they find the way to conserve the modularity of the membrane, i.e. the fact that each layer can be easily removed by the user, they become an excellent modular tool to timely respond to the fast changing needs in industrial processes. Beyond this principal technological aspect regarding the impact of the project, there is a more theoretical relevance: a new tool to analyze general membranes is made available to the scientific community, which, from now on, will be able to methodically evaluate also the smallest change in the characteristics of a membrane process, using cheap numerical computations, with a little effort in the implementation due to the principle of simplicity explained in section 2.2. Trying to couple macroscopic and microscopic physics through a mesoscale point of view (cf. Ocone, 2017) this is the tool that the theoretical and numerical part of the community in this field needs.

2.5 Reasons for the choice of host and co-host institution;

One of the fundamental aspects of the project is the use of a multi-scale technique where an important role is played by asymptotic analysis. The host supervisor has matured a long experience in asymptotic analysis and multiple scale expansions applied to NSE (cf. for instance, Meliga and Gallaire, 2011). In closer link with the present project, he treated multiphase and multiscale fluid transport phenomena in microscopic devices in Baroud *et al.*, (2010) and Zhu & Gallaire (2016). He will share with the PI his knowledge in asymptotics and numerics and through the host institution the necessary HPC resources to carry out the numerical results. The co-host supervisor, in some of his works (cf., for instance, Dölger *et al.*, 2014, Jensen *et al.*, 2016 and Rademaker *et al.*, 2017), demonstrates a strong expertise in transport phenomena, such as sugar transportation in plants, and with its physical background will provide important advices in the initial formulation of the hypothesis at the basis of the phenomenon, as stated in table 1.

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End of 10 page limit