

Computational Microfluidics for Geosciences

by

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Abstract

Computational microfluidics for geosciences is an emerging scientific discipline for investigating complex mechanisms in geological porous media. It allows for a fine-scale description of the coupled processes that occur in the soils and the subsurface employing advanced modelling tools (e.g. Computational Fluid Dynamics, Reactive Transport Modelling). In a scientific strategy relying on a cascade of scales nested within each other, computational microfluidics is, therefore, an essential tool to bridge the gap between spatial scales and guide the derivation of field-scale models rooted in a correct description of the elementary physical principles. It is a natural companion of lab-scale experiments in well-controlled environments by providing high-resolution mapping of pressure and velocity profiles, solute concentration, and mineral distribution that are not easily measurable experimentally. Computational microfluidics is also a powerful tool to perform sensitivity analysis for identifying the key parameters of the underlying processes or to explore ranges of conditions including pressure and temperature that are difficult to reach in the laboratory without dedicated equipment.

In this thesis, we review the state-of-the-art computational microfluidics as well as the author's contributions. They include pore-scale modelling of immiscible two-phase flow, mass transfer across interfaces, and geochemical processes (e.g. mineral dissolution/precipitation). We also introduce the concept of micro-continuum approaches for solving coupled processes in computational microfluidics and its usage for image-based simulations. The thesis outlines the open challenges, the current trends, and future directions in the field.

Résumé étendu

Les milieux poreux géologiques sont des objets d'étude fascinants. Une bonne compréhension des mécanismes d'écoulement et de transport au sein de ces matériaux est cruciale pour accompagner l'exploitation des formations souterraines à des fins énergétiques (géothermie, hydrogène orange, réservoirs pétrolifères), de stockage de gaz (dioxyde de carbone, hydrogène) et de déchets nucléaires, ou de gestion de la ressource en eau. A cause des échelles de temps et d'espace qui caractérisent ces projets – l'exploitation des ressources profondes fait appel à des puits de près d'un kilomètre de profondeur et les déchets nucléaires stockés dans les formations argilleuses ont vocation à y rester pour des milliers d'années – il est nécessaire de développer des capacités prédictives pour accompagner l'utilisation du sous-sol par l'homme tout en minimisant son empreinte environnementale.

La modélisation numérique et plus particulièrement la simulation des réservoirs est un outil d'aide à la décision pour les practiciens du sous-sol qui a comme objectifs d'estimer les ressources exploitables et permet d'envisager différents scénario d'ingénieurie (Chap. 1). Elle permet, par exemple, l'évaluation de la contamination des eaux souterraines, du devenir des contaminants, et la mise en place de méthodes efficaces pour leur dépollution qui constituent aujourd'hui un défi scientifique et sociétal majeur. De la même manière, la modélisation numérique permet d'envisager des stratégies variées pour étanchéifier, a priori, les barrières de confinement lors de stockages souterrains afin d'éviter les fuites vers la surface et d'assurer leur intégrité sur de longues périodes de temps. Cependant, la validité des modèles classiquement utilisés est restreinte à une petite gamme de paramètres qui ne correspond pas, le plus souvent, à la réalité des mécanismes impliqués. Les hétérogénéités des réservoirs, qu'ils soit poreux ou fracturés, complexifient fortement la mise en place d'une modélisation prédictive. Par ailleurs, les rétroactions entre l'écoulement diphasique, les transferts de masse aux interfaces et les processus géochimiques sont à ce jour peu explorées. Ainsi, l'estimation des quantités de dioxyde de carbone pouvant être séquestrées dans les formations souterraines afin de réduire la présence de ce gaz à effet de serre dans l'atmosphère nécessiste une connaissance de la physiques des écoulements multiphasiques, des mécanismes de piègeage capillaire au sein de la structure poreuse de la roche, des transferts de masse aux interfaces, et des intéractions fluide-roche. Ces physiques et leurs couplages mutuels sont mals (voire pas du tout) pris en compte dans les modèles actuels.

Cette thèse synthétise près d'une décennie de recherche sur la mise en place de modèles numériques multi-échelles pour simuler les phénomènes couplés dans les milieux poreux hétérogènes (Chap. 2). Elle est le fruit d'une collaboration soutenue avec des experts en microfluidique, en imagerie microtomographique, en ingénierie des réservoirs, en mécanique des fluides numériques, et en géochimie. Elle fait suite aux travaux successifs de l'auteur initiés à l'Institut de Mécanique des Fluides

de Toulouse sur le changement d'échelle des phénomènes de transfert de masse en régime diphasique dans les doubles milieux (2009-2012), puis sur la modélisation de la turbulence quantique dans les systèmes poreux (2013). A partir de 2013, ces travaux s'orientent vers l'étude des milieux géologiques. A Stanford University (2013-2018), ils portent sur l'étude des mécanismes impliqués, à l'échelle du pore, dans les procédés de stockage de CO_2 dans les aquifères salins profonds (2013-2018), et depuis 2018 (BRGM, CNRS) sur le développement de modélisations numériques couplées hydro-géochimiques à différentes échelles d'intérêt allant du nanomètre au kilomètre.

La stratégie scientifique, pour développer des modèles prédictifs basés sur les principes physiques élementaires, utilise une modèlisation en cascade d'échelles imbriquées les unes dans les autres (Chap. 4). L'apparition d'approches microfluidiques pour les géosciences, l'amélioration des techniques d'imagerie ainsi que le développement croissant des puissances de calcul permettent aujourd'hui d'étudier les processus à l'échelle du pore et du réseau de pores où les modèles physiques sont mieux établis. Ainsi, la microfluidique offre un cadre unique pour visualiser les couplages hydro-géochimiques avec une haute résolution spatiale et temporelle en réalisant des expériences dans des environnements contrôlés. En parallèle de cet effort expérimental, la modélisation numérique est également en plein essor. Les approches dites *Digital Rock Physics* résolvent la physique directement dans des images en trois dimensions de roches digitalisées à l'aide d'outil comme la microtomographie, afin de caractériser, par simulation, les propriétés pétrophysiques de la roche.

La modélisation des phénomènes couplés à l'échelle du pore est le point de départ de la microfluidique numérique pour les géosciences, le moteur physique de la *Digital Rock Physics* (Chap. 5). Ce concept, développé par l'auteur et ses collaborateurs, est la contrepartie digitale des expériences en laboratoire. La microfluidique numérique augmente l'expérience en laboratoire en fournissant, notamment, des cartographies dynamiques des champs de vitesse, de pression, de température et des concentrations en phase aqueuse, ainsi que la distribution des phases minérales, données qui ne sont pas aisément mesurables expérimentalement. C'est également un outil puissant pour effectuer des études de sensitivité afin d'identifier les paramètres clés régissant les processus, ainsi que pour explorer des gammes de pression et température difficilement atteignables en laboratoire sans un équipement dédié. Les modèles actuels sont capables de simuler des écoulements multiphasiques, des phénomènes de solubilisation aux interfaces, ainsi que des processus géochimiques à la surface des minéraux (e.g. dissolution, précipitation). Bien que cette discipline récente en soit encore à ses balbutiements, la microfluidique numérique a déjà apporté des résultats tangibles comme l'estimation des surfaces réactives accessibles en fonction des conditions d'écoulement lors des processus géochimiques, ou encore la mise en évidence de barrières gazeuses changeant la dynamique de formation des instabilités de dissolution. Cette thèse présente un état de l'art de la microfluidique numérique, de ses verrous scientifiques, et des tendances actuelles et futures.

Les approches micro-continues (Chap. 6) développées par l'auteur et ses collaborateurs permettent de résoudre les physiques couplées qui sont au coeur de la microfluidique numérique pour les géosciences. Ces approches sont basées sur l'équation de Darcy-Brinkman-Stokes (Chap. 3) qui a la capacité de modéliser les écoulements dans les systèmes possédant deux tailles de pore caractéristiques. Elles

sont par exemple un candidat idéal pour décrire le transport dans les systèmes poreux fracturés où le réseau de fractures est représenté explicitement alors que la matrice est représentée par un milieu homogénéisé où la structure poreuse est décrite par des concepts tels que la porosité, la perméabilité et la surface spécifique. Les approches micro-continues pour l'échelle du pore ont initialement été mise en place pour prendre en compte les détails de la géométrie poreuse plus petits que la résolution de l'imagerie tomographique et donc non-visible sur les images. L'objectif était d'inclure l'impact de la porosité non observale dans le calcul de perméabilité absolue à partir de simulations multi-échelles d'écoulement monophasique. Depuis cette vocation première, le concept n'a cessé de s'étendre et les modèles actuels sont capables de décrire les écoulements diphasiques en présence de microporosité, de simuler les processus hydro-géochimiques comme la dissolution et la précipitation des minéraux ou encore la déformation mécanique de la matrice solide comme le gonflement des argiles. Loin de rester cantonnées aux petites échelles, les approches micro-continues sont intrinsèquement multi-échelles et présentent l'avantage considérable de simuler les processus couplés également à l'échelle du terrain. Ainsi, un seul et même système d'équations permet de simuler les processus couplés dans les milieux poreux sur des échelles caractéristiques allant du nanomètre au kilomètre et au delà.

Fervent supporteur de la Science ouverte, l'auteur met ses codes en license libre à disposition du plus grand nombre. *porousMedia4Foam* est le companion digital incontournable de cette thèse (Chap. 7). Cet outil numérique, qui se présente sous la forme d'une librairie open-source pour la plateforme de simulation OpenFOAM®, est développé autour des approches micro-continues. Il permet de modéliser les phénomènes couplés hydro-géochimiques à des échelles d'intérêt allant de celle du réseau de pores à celle du terrain en passant par l'échelle de la colonne en laboratoire. L'utilisation de cette librairie va bien au-delà du domaine des géosciences et son application concerne toute étude faisant intervenir des matériaux poreux qu'ils soient naturels ou manufacturés (e.g. contacteurs gaz-liquide en génie chimique, tablettes effervescentes dans l'industrie pharmaceutique, tissus biologiques, marc de café, piles à combustible...). Le développement de *porousMedia4Foam* est continu et inclu les contributions passées, présentes et futures des doctorants, post-doctorants, et partenaires associés à ces recherches.

Pour mes futures lignes de recherche, je souhaiterai comprendre comment les particules colloïdales (e.g. nanoparticules métalliques, bactéries) contrôlent les écoulements diphasiques dans les milieux poreux. Déchiffrer ces mécanismes complexes est fondamental pour développer de nouvelles technologies de remédiation des aquifères contaminés par des solvents chlorés et des hydrocarbures. Cette sorte de pollution est reconnue comme l'une des plus difficiles à éliminer car des gouttelettes de contaminants sont piégées par les forces capillaires dans l'espace poral de la formation. La stratégie scientifique présentée dans ce mémoire permettra une connaissance approfondie, à l'échelle du pore, des processus capables de remobiliser les gouttes ainsi piégées. Des approches de microfluidique numérique dédiées sont un atout considérable pour guider la mise en place de nouvelles stratégies de remédiation. Leur développement requiert la poursuite des efforts qui m'occupent depuis près d'une décennie.

Acknowledgements

Ten years have passed since I defended my PhD thesis. I was thinking of great achievement at that time. Now that I am looking back at all the work done since then and all the people met all around the world, I realize that I was just at the beginning of my long trip into science. And the best is yet to come!

The body of research presented in this document results from research based on different projects, and I would like to thank all the people that I met during my stay at the Institut de Mécanique des Fluides de Toulouse, at Stanford University, and now at the Institut des Sciences de la Terre d'Orléans. It will be too long to list all the people that crossed my path and with whom I shared good times, knowledge, and science problems. Your friendship means a lot to me. Nevertheless, I am deeply indebted to Michel Quintard who ignited my curiosity for porous media, and Hamdi Tchelepi who upscaled my view of managing research. I learned a lot.

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Ten years of research is also ten years spent with Sophie in the lab and at home, and now with Marianne. This work could not have been done without them.

Best,
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Chapter 1

Introduction

Geological porous media are fascinating objects. They play a major role in the transport of mass in the soils and the subsurface. The consequences of their exploitation, however, are challenging to predict because of the multi-scale and multi-physics nature of the processes at stake. Numerical simulation is a real asset to understand the coupling between processes, and to guide the exploitation of natural resources while minimizing the footprint of anthropogenic activities.

1.1 Effective use of the subsurface for resources and energy applications

Effective use of the Earth subsurface for resources and energy applications requires very good control of time- and space-dependent fluid flow regimes. Such applications include the exploitation of groundwater reservoirs, the storage of carbon dioxide (CO_2), hydrogen (H_2), or nuclear waste in deep geological formations for thousands of years, the development of geothermal energy, and the recovery of oil and gas (DePaolo and Cole 2013). In this section, we use the example of CO_2 sequestration in the subsurface – a research topic familiar to the author – to briefly illustrate the challenges associated with the exploitation of underground resources.

Processes involving storage into the subsurface heavily rely on the presence of impervious geological barriers. Large-scale deployments of geologic storage (carbon dioxide, hydrogen, nuclear wastes) require minimizing the risk of leaks towards groundwater and the surface (Fig. 1.1). Fractures, fissures, and natural faults are discontinuities that present an important risk of leakage. By changing the equilibrium conditions of the brine, the injection of fluids in the subsurface can lead to an enlargement of the flow pathways in the cap-rock by dissolving rock minerals, and therefore significantly alters the cap-rock impermeability. An in-depth understanding of the dissolution and precipitation of minerals in porous and fractured porous media and the complex feedback on the transport of fluids is essential to characterize fully these reactive systems and to design efficient sealing strategies.

The effective carbon dioxide storage capacity worldwide is uncertain because the influence of complex coupled processes occurring during and after injection is often neglected. During the process of CO_2 storage by residual or solubility trapping, capillary forces are dominant. These forces are actually involved in all the subsurface processes that include two or more fluid phases, such as flow in the unsaturated zone

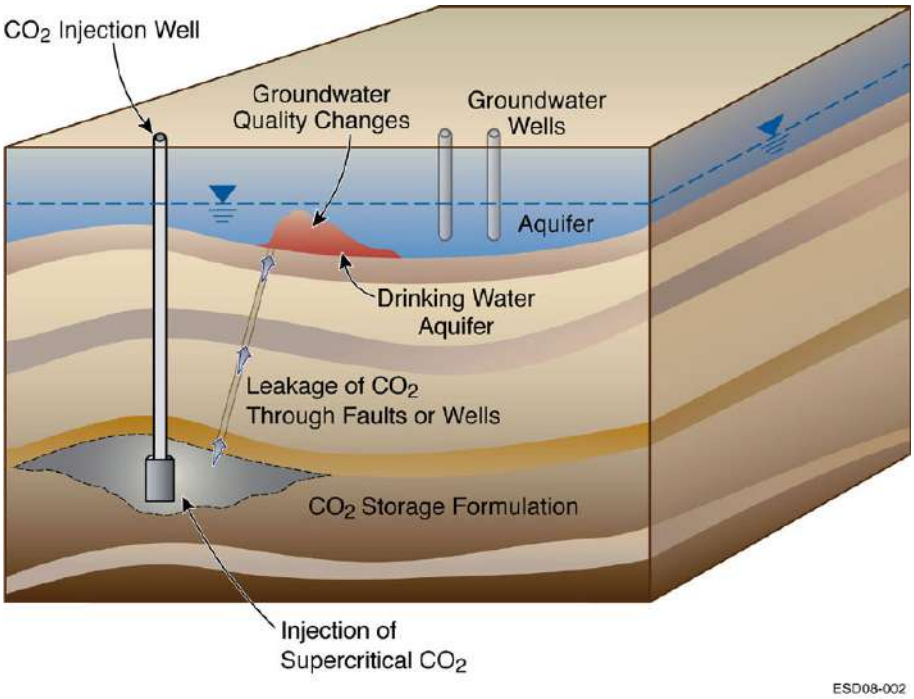


Figure 1.1: Illustration of CO_2 storage into the subsurface. Leaks in the cap-rock can contaminate fresh water and flow to the surface (source: Lawrence Berkeley National Laboratory).

or groundwaters contaminated with hydrocarbon and chlorinated solvents. Capillary effects are a desired mechanism to store large quantities of CO_2 in geological formations, or on the contrary they are a barrier that has to be overcame to remobilize contaminant trapped in soils and aquifers. The success of a secure and permanent underground storage of CO_2 and of an efficient environmental remediation depends largely on our understanding of two-phase fluid displacements leading to residual saturation, and solubilization into formation water.

Likewise, interphase mass transfer is either a desired mechanism to solubilize CO_2 in brine or one to avoid when harmful chemicals are released from contaminants to groundwater. In these multi-fluid flow problems, a complex interplay of physicochemical factors controls the fluid displacements, trapping, and remobilization. It includes the fluid pair properties (e.g. density and viscosity ratio, interfacial tension, fluid composition, solubility, molecular diffusivity), the matrix properties (e.g. permeability, pore size distribution, wettability of minerals), as well as the flow rates. Moreover, these factors change over time because of the transfer of species across the interface separating different phases. Interphase mass transfers have different consequences ranging from a simple change in the fluid composition to stronger impacts on the flow properties such as wettability alteration, surface tension reduction, or Ostwald ripening, thus leading to enhanced residual trapping or on the contrary to remobilization of trapped phases.

The deployment of new exploitation or remediation technologies is often a very long route starting in laboratories with small-scale experiments to pilots in the field. It is not rare for it to take a couple of decades before a new concept is sufficiently developed to allow its daily usage in the field. Hence, any solution offering to shorten the development time is a real asset for engineering.

1.2 Objectives and challenges

1.2.1 Development of predictive capabilities

Field trials are costly and time-consuming to carry out and are currently a barrier to transitioning a new energy or storage concept from a lab-scale process to a practical field-scale deployable technology. Numerical modelling, and reservoir simulation in particular, allows for predicting field performance and ultimate recovery for various field development scenarios to evaluate the effects on recovery of different operational conditions and compare the economics of different recovery methods. They are also key in the assessment of storage capacities (e.g. CO_2, H_2) and their long-term integrity over thousands of years. In water resources management, they are used to evaluate the fate of contaminants in aquifers. Reservoir simulation is, therefore, the ideal tool to identify the origin of pollution and to try new remediation strategies. The exploitation of geothermal, groundwater, and petroleum reservoirs is greatly guided by reservoir simulation as it also allows assessments of multiple development alternatives including production scenarios and optimization of wells placement.

Reservoir simulation relies on the construction and operation of three-dimensional models whose appearance and behaviour represent the actual reservoirs (Fig. 1.2). The simulators are based on a set of partial differential equations that describes the physical processes active in the reservoir. Of course, a mathematical model lacks the complex reality of a reservoir and is always subject to certain assumptions. This means that a reservoir simulation is only valid for a certain range of parameters. In many cases, however, the range of validity of standard models is not representative of the processes involved in the subsurface. As of today, the state-of-the-art simulators are not able to model accurately the complex feedback between multiphase flow, interfacial mass transfer, and fluid-mineral interactions when the system equilibrium conditions are displaced due to anthropogenic activities. This is a real concern for applications such as CO_2 storage or aquifer remediation as the mechanisms leading to capillary trapping within the porous matrix as well as the geochemical processes producing leakage pathways toward the surface are poorly described. It is, therefore, a necessity to develop reservoir models that consider coupled processes occurring in a multi-scale environment. The ideal simulator has to be rooted in elementary physical principles. It has to account for the change of rock properties due to mechanical and geochemical processes, for wettability alteration to assess the storage capacity of a reservoir over long periods, and for reservoir spatial heterogeneities including fractures, cracks, and faults.

1.2.2 Open challenges

The development of robust and efficient field-scale simulators is not an easy task. The following list contains several sets of scientific and technical challenges that limit the development of predictive capabilities for subsurface processes.

A multi-physics problem The processes involved in geological porous media relate to different physics (e.g. hydrodynamics, electrostatics, geochemistry, mechanics) that are strongly coupled to each other. A detailed understanding of the complex feedback between the processes is crucial to quantify the relevant couplings and assess the dominant mechanisms.

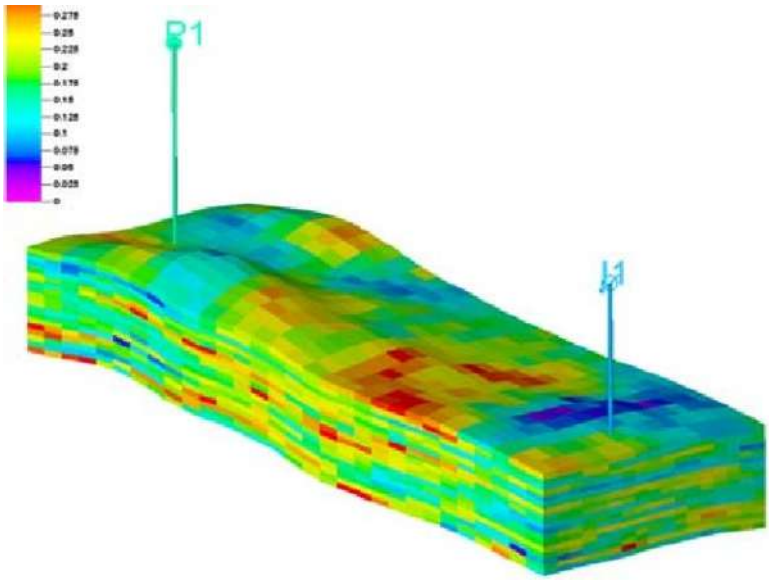


Figure 1.2: Geometry of a shallow marine North Sea conceptual model including the grid, the porosity field, the depth and the well positions after Akervoll and Eirik Bergmo (2010). The standard physical laws used in reservoir simulations have very often ranges of validity that are not representative of the actual processes.

A multi-scale problem Transport phenomena in heterogeneous geological formations are a multi-scale problem with characteristic length scales ranging from the molecular level to the scale of the reservoir. The presence of fractures, cracks, and faults complicates the modelling by the introduction of additional heterogeneity and strong anisotropy. They are discontinuities within the porous matrix that favor fluid migration and solute transport.

Computational challenges The development of efficient and reliable numerical models at the various scales of interest is challenging. In particular, the models have to be verified against well-controlled experiments before being used with confidence for prediction. Moreover, the large computational domains required to be representative of real-life demands High-Performance Computing.

Experimental and visualization challenges The opacity of natural porous media makes the visualization of subsurface processes and underlying mechanisms particularly challenging. Due to the heterogeneous nature of rocks, it is challenging to track the underlying processes efficiently in the pore-space. Standard characterisation techniques often rely on indirect macroscopic measurements that require inverting a model to derive the rock properties.

Identification of key parameters The flow physics involved in transport in sedimentary rocks is quite complex. For example, the transport, deposition and aggregation of particles, as well as the fluid remobilization depend on a large number of parameters including pH, salinity, flow rate, fluid physical properties, interfacial tension, wettability, surface roughness, surface charges, heterogeneity of the rock microstructure, particle size distribution, particle concentration, and particle coating.

Moreover, modification of one parameter can generate a highly non-linear response of the system leading to challenging interpretations of the results that are very often qualified as controversial and inconsistent in the literature (Donath et al. 2019).

1.2.3 The interest in pore-scale physics

To obtain deeper insight into the physics of flow and transport in natural media, there has been growing interest to start at the pore scale, where the physics is better understood and to then upscale the results to a macroscopic Darcy-like representation. At the pore scale, the spatial distribution of the constituents (e.g., sand grains) and the geometry of the space enclosed by the solid skeleton can be described in great detail. This recent interest is driven by the enormous growth in computational capabilities, the accelerating improvement in imaging techniques, and by the development of microfluidic devices for geosciences. The Porous Media Research Group at the Earth Sciences Institute of Orléans (ISTO) is investing substantial efforts in pore-scale research to decipher the complex coupled physics in geological porous materials using a combination of microfluidic experiments, numerical simulations, and high-resolution imaging.

This thesis is dedicated to *computational microfluidics for geosciences* that is the numerical part of this triptych of methods. It allows for solving coupled processes including hydro-bio-geochemical processes at the pore-scale. Computational microfluidics is therefore an essential tool to complement and augment laboratory experiments in well-controlled environments by providing maps of velocity, pressure, temperature, and concentration that are not easily measurable in the lab. Its ability to perform sensitivity analysis for identifying the key parameters governing the coupled processes as well as simulations in pressure and temperature conditions that cannot be achieved in the lab without dedicated instruments is an invaluable asset of the technique.

Importantly, with the recent and accelerating improvements in imaging techniques, such as X-ray computed micro-tomography, it is now doable to construct high-resolution three-dimensional (3D) descriptions of rock samples and to use computational microfluidics combined with high-performance computing techniques to solve the flow and transport equations in the pore space (Blunt et al. 2013; Wildenschild and Sheppard 2013). This kind of image-based simulation is sometimes referred to as *Digital Rock Physics*. For example, it is now somewhat routine to estimate the absolute permeability and dispersion tensors by solving the Navier–Stokes equations using high-resolution images of rock samples (Andrä et al. 2013a,b; Arns et al. 2005; Blunt et al. 2013; Guibert et al. 2015; Kainourgiakis et al. 2005; Soulaine et al. 2021a, 2016; Spanne et al. 1994). Fig. 1.3 illustrates Digital Rock Physics results.

The rock properties that can be evaluated using *Digital Rock Physics* depend strongly on the capabilities of computational microfluidics to solve multi-physics and coupled processes. The field is still in its infancy and is growing rapidly. Cutting-edge computational microfluidics handle hydro-geochemical processes, two-phase flow, charge displacement, solid deformation, and even biofilm growth. There is still, however, a lot of improvements and verification to develop computational microfluidics valid over a wide range of parameters. Moreover, even though pore-scale flow simulators can now handle up to billion-cell representations (Trebotich

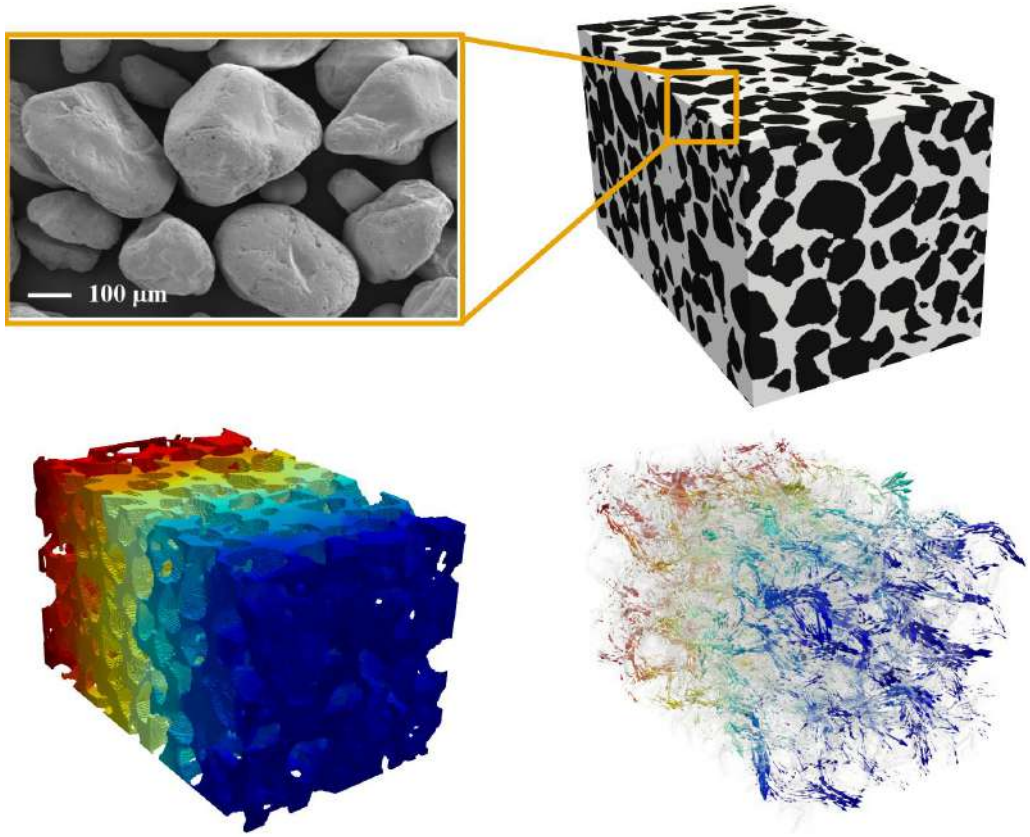


Figure 1.3: Image-based simulation in a sandpack *(top left)* after Soulaine et al. (2021a). The sandpack has be imaged *(top right)* using ISTO's Nanotom microtomograph 180NF Phoenix X-Ray. The absolute permeability is simply obtained by averaging the pressure *(bottom left)* and flow *(bottom right)* profiles and inverting Darcy's law.

and Graves 2015), the actual physical scale of the rock sample is a few cubic millimeters. This is because natural porous media are intrinsically multiscale. In many formations, complex spatial arrangements of the constituents, including the pore space, are observed all the way down to the nanometer scale. So, using computational microfluidics for all scales all the time is simply impossible and pore-scale modelling is only an intermediate step towards the development of robust field-scale models.

1.3 Outline of the thesis

This thesis summarizes the research I have been doing for almost a decade to decipher the complex flow and transport mechanisms occurring in the soils and the subsurface. The dissertation is organized as follows. Chapter 2 gives an overview of my research profile, my curriculum vitae, and my investment in the scientific community. In Chapter 3, I discuss the different scales of the description of flow and transport in porous media, and I show that Darcy's law is no longer an empirical law but a sound model rooted in the elementary physical principles by averaging pore-scale physics. Actually, the averaging procedure results in the Darcy-Brinkman-Stokes equation that is the cornerstone of the multi-scale approaches proposed in

this thesis. Then, in Chapter 4, I present the scientific strategy I adopt in most of my research to develop predictive capabilities for simulating coupled processes in porous systems. It relies on a multi-scale approach centered on a fine-scale description of hydro-chemical processes using a combination of numerical simulations, microfluidic experiments, and high-resolution imaging. Chapter 5 is devoted to computational microfluidics for geosciences with an emphasis on my own developments in terms of multiphase flow and reactive transport. In Chapter 6, I review the concept of micro-continuum models –an important pillar of my research– for solving coupled systems both in terms of multi-physics and multi-scale modelling. In Chapter 7, I introduce *porousMedia4Foam*, an open-source package that is the digital companion of the present manuscript. Finally, I close with conclusions (Chapter 8) and longer-term perspectives.

Chapter 2

Research Profile

I am trained both as a computational scientist and a fluid dynamicist with a strong interest in flow and transport in porous media. My research is driven by the desire to understand and describe the transport mechanisms in porous materials at different length and time scales. I am looking at a large variety of complex physics in porous media and I want to understand how the small-scale events (down to nanometers or micrometers) affect the phenomena at larger scales (up to the column- or field-scale). My approach to bridge the gap between the scales consists in developing the relevant physical descriptions of the phenomena in a logic of a cascade of scales nested within each other and to find how the spatial scales are linked between them. My research follows a rigorous approach to derive large-scale models rooted in pore-scale physics using consistent averaging techniques and efficient numerical algorithms.

2.1 Research background

2.1.1 Context of research

Since my early works, my research is motivated by the desire to understand and describe the transport mechanisms in porous materials over a broad spectrum of spatial and temporal scales. They can be manufactured (fuel cells, chemical contactors, heat exchangers, cements) or natural (geological media, biological tissues, canopy). I am looking at a large variety of complex coupled physics in porous media including inertial and turbulent effects, multiphase flows (thin films, surface tension dominated flow), mass transfer across interfaces, reactive transport with an evolution of the pore-space morphology (dissolution or precipitation at the mineral surfaces) and more exotic topics such as quantum turbulence in superfluids. A common challenge to model these phenomena is the material heterogeneity that can span a wide range of length scales. In particular, I want to understand how the small-scale events (down to nano or micrometers) affect the transport phenomena at larger scales (up to the column or field scale). My approach to bridge the gap between the scales consists in developing the relevant physics in a logic of a cascade of scales nested within each other and to find how the spatial scales are linked between them. My research follows a rigorous approach to derive large-scale models based on Darcy's law or variants (usually the scale of interest for engineering applications) from pore-scale physics studied with an integrated usage of Computational Fluid Dynamics, microfluidic experiments and micro-tomography imaging, as well

as using consistent homogenization techniques and efficient numerical algorithms.

2.1.2 Chronology and research topics

This section summarizes my past experience in research from my Master degree in 2008 to nowadays. Fig. 2.1 is a graphical abstract of my curriculum vitae.

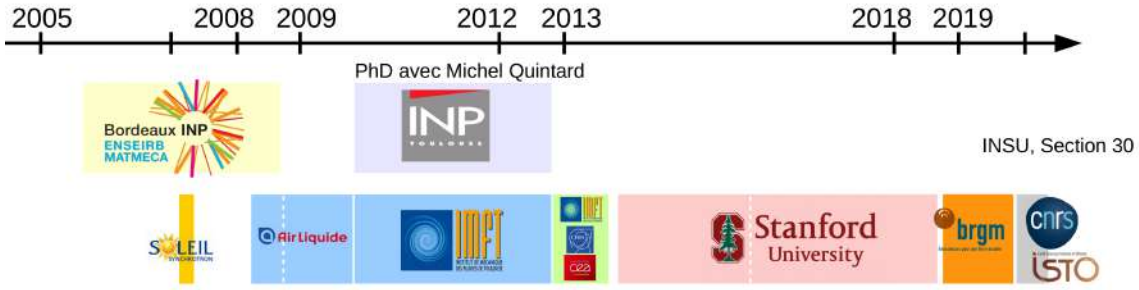


Figure 2.1: Graphical abstract of Cyprien Soulaine's curriculum vitae.

I am trained as a computational scientist with a major in fluid dynamics. I worked 18 months (including a 6-months internship) as a Research Engineer at Air Liquid Research Center where I developed dual-porosity models coupling heat and mass transfer in adsorption-based gas separation columns.

Eager to improve my understanding of the gap between the different scales in porous media, I did a Ph.D. at the Institut de Mécanique des Fluides de Toulouse (IMFT) under the supervision of Pr. Michel Quintard. I obtained an industrial fellowship (CIFRE) sponsored by Air Liquide to investigate gas-liquid flow in cryogenic air distillation columns equipped with structured packing. The same kind of columns is also used in carbon capture technologies.

I continued my research on porous media flow by a 9-months postdoc at IMFT co-funded by IMFT, the European Organization for Nuclear Research (CERN), and the Commissariat à l'Energie Atomique (CEA) to investigate instabilities in superfluid helium flow through the superconducting porous magnets of the Large Hadron Collider.

In October 2013, I joined the group of Prof. Hamdi Tchelepi in the School of Earth, Energy, and Environmental Sciences at Stanford University, first as a postdoc then as a Research Associate. The group's expertise lies in developing efficient simulators at the reservoir-scale. I have started and led a new research activity centered on pore-scale modelling with the final objective to improve the physical models implemented in reservoir simulators. This includes applications such as CO_2 storage in deep saline aquifers, geothermal energy, and unconventional reservoirs. To that end, I have developed an advanced Digital Rock Physics[1] research program covering different modelling steps, from the design of new models to their validation by comparison with experiments. It was funded by the US Department of Energy and a consortium of industrial partners. This experience led me to the investigation of coupled two-phase flow and reactive transport in geological formations.

[1]Digital Rock Physics: Characterization of rock properties using simulations in three-dimensional images

In September 2018, I joined the French Geological Survey (BRGM) as a Research Scientist in Francis Claret's team in the Department of Water, Environment, and Ecotechnologies. I continued my research on modelling coupled processes in geological porous media with the objective to assess the long-term effectiveness and the environmental impact of geological storage.

I permanently secured my research activities by entering the CNRS[2] in October 2019. I integrated the Earth Sciences Institute of Orléans (ISTO) – a joint research laboratory with CNRS, Université of Orléans, and BRGM – with the objective to predict the transport of contaminants in aquifers, to assist the design of efficient remediation technologies and to assess the long-term integrity of storage systems in the subsurface as well as the safe exploitation of subsurface energy resources. I am now the Head of the Porous Media Research Group whose ambition is to develop a leading-edge scientific centre of excellence on flow and transport modelling in geological systems by developing cutting-edge multi-scale simulators along with columns and microfluidics experiments.

2.1.3 Research collaboration and scientific network

Thanks to my participation to large-scale projects I had the opportunity to develop an important network of collaborators both at the national and international level. For example, I was part of the Center for Nanoscale Control on Geologic CO_2 that involved 7 major US research institutions including Lawrence Berkeley National Laboratory (LBNL), the Oak Ridge National Laboratory (ORNL), Ohio State University, Princeton University, Purdue University, Stanford University, and Washington University in St. Louis. Overall, I have more than 52 co-authors. Most of the time, these scientific collaborations bring experiments (lab-scale or field-scale) to complement my computational modelling.

I am now an active and recognized scientist in the emerging area of Digital Rock Physics, and in recognition of the new generation of reactive multi-scale models I am developing, I have been invited to present my work in major international conferences in my field including the prestigious Gordon Research Conference on Permeable Media in 2018. The micro-continuum approach is now more and more used in the porous media community (see Chapter 6).

2.1.4 Investment in the scientific community

I am thoroughly invested in the scientific community on flow and transport in porous media. I organized minisymposia in international conferences (e.g., SIAM Geosciences 2015, CMWR 2016, Interpore 2018). I am also a regular reviewer for scientific journals including Transport in Porous Media, Advances in Water Resources, Journal of Hydrology, Journal of Fluid Mechanics, and Water Resources Research.

Moreover, I have always been a fervent supporter of open science, and I release my codes in open-source. Most of my code developments rely on OpenFOAM[3], an open-source library for solving partial differential equations on unstructured grids using the finite-volume method. I am involved in FOAM-U[4], the association of

[2] CNRS: French National Centre for Scientific Research

[3] https://www.openfoam.org

[4] https://www.foam-u.fr

the OpenFOAM users in France. I have been elected president of the association in July 2020. FOAM-U aims at bringing the different French OpenFOAM players together through an annual meeting (60-100 participants). I also organize short courses on open-source computational fluid mechanics using OpenFOAM in various places worldwide including universities and research laboratories (e.g. Institut de Mécanique des Fluides de Toulouse, Géosciences Montpellier, Stanford University, Institut des Sciences de la Terre d'Orléans, Beijing Normal University), industries (e.g. BRGM, Air Liquide), international conferences (e.g. Interpore 2016 in Rotterdam, Interpore 2020 Virtual), summer's school (e.g. 4th and 5th Cargèse Summer School on flow and transport in porous and fractured media), and virtually. Overall, I have trained more than 300 students, engineers, and scientists. Besides the obvious academic aspect, these short courses help me to promote the libraries I develop and to initiate new collaborations.

2.2 Current and past achievements

In this section, I briefly describe my current and past achievements. More details regarding the notion of scales in porous media, the scientific strategy I adopt, and the pore-scale physics I am investigating are found in the next chapters.

2.2.1 Injection and sequestration of CO_2 into the subsurface

Capture and geological storage of CO_2 to reduce carbon emission is among the most critical energy technologies of the next century. My research in the group of Pr. Hamdi Tchelepi in the Department of Energy Resources Engineering at Stanford University was supported by the US Department of Energy through the Center for Nanoscale Controls on Geologic CO_2. The objective was to improve the fundamental understanding of the pore-scale processes associated with the injection and sequestration of CO_2 into the subsurface and to assess the long-term issues such as the efficiency of CO_2 retention in reservoir rocks resulting from capillary and dissolution trapping, and from the conversion of dissolved CO_2 to solid carbonate. Interfacial properties and dynamics control fluid flow patterns and trapping by their influence on wetting properties, multiphase fluid and chemical transport, and reactivity. I developed computational tools to simulate these different processes at the pore-scale including the effect of sub-voxel porosity in X-ray imaging (Soulaine et al. 2016), capillary trapping (Abu AlSaud et al. 2017; Roman et al. 2016), the transfer of CO_2 from the supercritical phase to the brine (Graveleau et al. 2017; Maes and Soulaine 2018), the dissolution of a calcite crystal (Soulaine et al. 2017b, 2018), and the emergence of wormholes (Soulaine and Tchelepi 2016). The numerical models were validated using microfluidic experiments.

2.2.2 Digital Rock Physics

With the recent and accelerating improvements in imaging techniques, such as X-ray computed microtomography combined with High-Performance Computing techniques, it is now possible to have high-resolution three-dimensional descriptions of rock samples and to solve Navier-Stokes-based simulation of flow and transport in

the pore space. The use of images of the pore space combined with dynamic modelling is sometimes referred to as Digital Rock Physics (DRP). DRP allows a better characterization of the detailed structures of rocks and a deeper insight into the rock-fluid interactions. I develop simulation tools for DRP that solve complex physics at the pore-scale including multiphase flow and reactive transport processes. DRP aims at characterizing rock properties including absolute and relative permeabilities, capillary pressure curves, dispersion tensors, and permeability-porosity relationship.

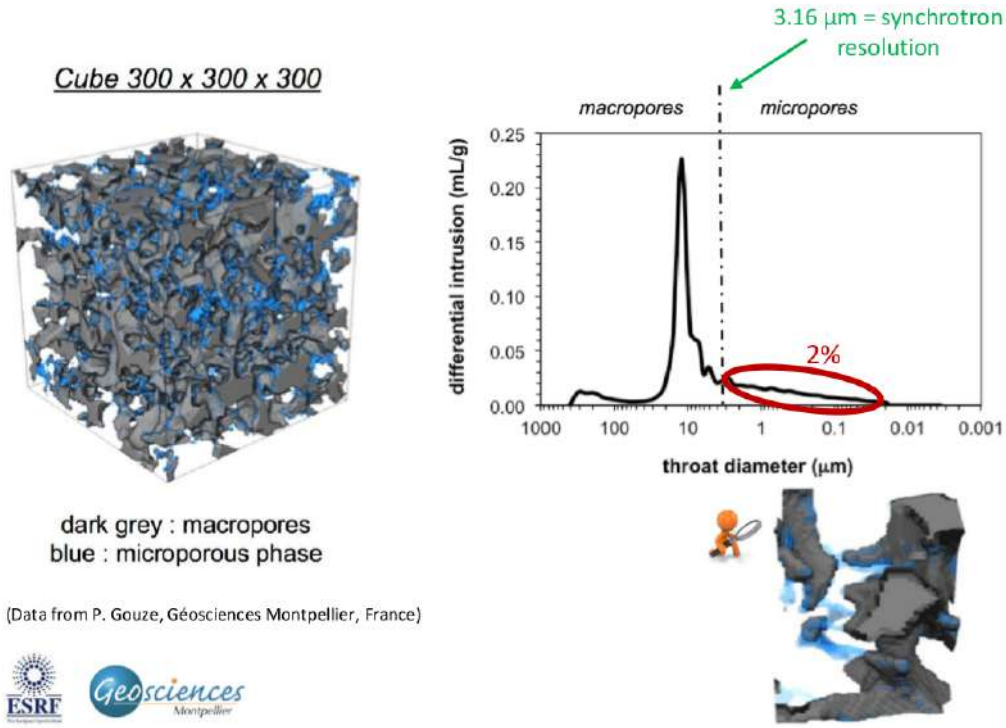


Figure 2.2: Pore space of a Berea sandstone imaged at the European Synchrotron Radiation Facility (ESRF). About 2% of the pores have throat diameter smaller than the imaging instrument resolution. This subvoxel porosity is not visible in the images but cannot be neglected because it participates to the medium connectivity (adapted from Soulaine et al. (2016))

The originality of my approaches with respect to other DRP is that I consider the pores that are too small to be visible in the images but do participate in the flow connectivity. For example, we proposed a filtering approach to compute absolute permeability in a three-dimensional image of Berea sandstone that contains up to 2% of the pores smaller than the imaging instrument resolution (Soulaine et al. 2016). We show that neglecting this subvoxel porosity could yield in a misestimation of about 100 % (see Fig. 2.2). If the subvoxel pore-throat size is nanometer or subnanometer, as in shales, the continuum mechanics breaks down and molecules transport include slip effects, adsorption, and Knudsen diffusion. In Soulaine et al. (2019), I extended the filtering approach using subgrid models for nanoporous media to account for nanoscale effects. Recently, we developed subgrid models for two-phase flow that were used to assess the rock relative permeabilities in the presence of subvoxel porosity (Carrillo et al. 2021).

2.2.3 Multi-scale modeling of gas-liquid flow in distillation columns

My Ph.D. research at the Institute of Fluid Mechanics of Toulouse (IMFT), supervised by Pr. Michel Quintard and funded by Air Liquide, concerns the modelling of cryogenics air distillation columns equipped with structured packings (Soulaine 2012). Such structures – illustrated in Fig. 2.3 – are made of the assembly of metal corrugated sheets. Structured packings maximize the exchange surface between gas and liquid while pressure drops remain low. I developed a comprehensive model rooted in pore-scale physics using a multi-scale analysis to simulate gas-liquid flow and species transport through the distillation columns. My research focused on three main points: *(i)* the effect of turbulence in porous media, *(ii)* the spreading of liquid films in structured packings, *(iii)* and the multi-component mass transfer in unsaturated porous media.

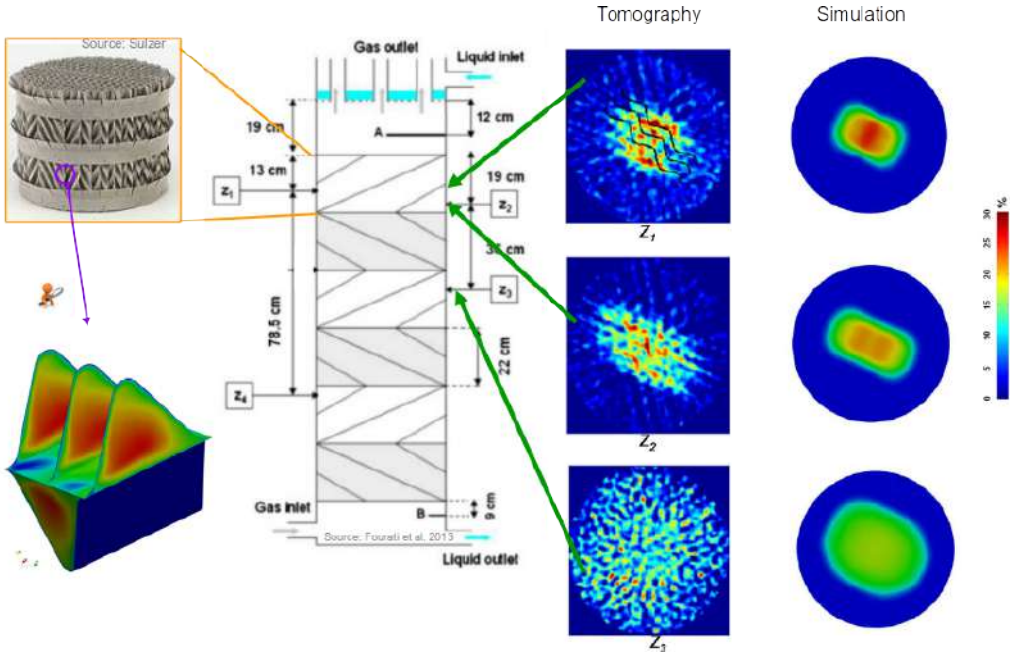


Figure 2.3: Multi-scale analysis of two-phase flow in distillation columns equipped with structured packings

First, a non-Darcean law that includes inertial and turbulence effects has been derived using homogenization techniques (Soulaine and Quintard 2014). Turbulence modelling in porous media is still an open challenge. Existing models use time and space averaging operators to derive turbulence models in porous media along the same lines as the standard Reynolds-Averaged Navier-Stokes (RANS) equations used in Computational Fluid Dynamics (de Lemos 2012). The theory we developed using the method of volume averaging demonstrates that under the assumption of scale separation, turbulent flows in porous media can be described using an extended Darcy-Forchheimer law which is much simpler than RANS models for porous media. Our approach computes macroscopic parameters from turbulent simulations at the pore-scale. Moreover, our Darcy-Forchheimer law handles the microstructure anisotropy and it is in agreement with empirical laws used in the chemical engineering literature.

Then, to model the liquid spreading in structured packing, both liquid films, one-per-sheet, were treated separately and upscaled as multi-continua (Soulaine et al. 2013, 2014a). I have identified the mechanisms that lead to the liquid radial dispersion effects: the main part comes from the geometry itself, the other part is due to the capillary effects at the contact points between adjacent sheets that result in a local redistribution of the liquid from one sheet to the other. The numerical model I proposed accounts for these different mechanisms and simulation results of the liquid distribution are in very good agreement with experimental data acquired by tomography imaging within a lab-scale column at IFP Energie Nouvelles during the Ph.D. of Manel Fourati (Soulaine et al. 2014a).

Finally, I investigated the transport of chemical species and their hydrodynamic dispersion in structured packings under two-phase flow conditions. Using homogenization techniques, I have developed a column-scale model that describes the multi-component mass transfer across the immiscible interface that separates the gas and liquid phases. The novelty of this work is the consideration of the interface velocity in the upscaling process (Soulaine et al. 2011). I continue to work on this research topic in different contexts by looking at the pore-scale dynamic mass transfer processes in two-phase flow by developing the Continuous Species Transfer modelling technique described in Chapter 5.

The associated computational tools are now actively used in Air Liquide to design enhanced packings. This research was further pursued at IMFT by Sylvain Pasquier during his Ph.D. (Pasquier 2017). Actually, the methodologies and theories developed during my Ph.D. go way beyond the modelling of processes in distillation columns equipped with structured packings. Indeed, because of their generic aspect, they also apply to all kinds of porous materials including manufactured (packings, membrane, fuel cells...) or natural (geological formation, karst systems, fractured media, biological tissue...) porous media.

My Ph.D. research has been rewarded by two academic awards:

- Léopold Escande award for outstanding Ph.D. thesis (less than 10% of the thesis defended in 2012) from National Polytechnic Institute of Toulouse,
- Jean Nougaro award for the best thesis in Engineering from Toulouse Academy of Sciences, France (2013).

2.2.4 Quantum turbulence in porous media

Below 2.17 K, helium no longer behaves as a classical fluid and transitions to a superfluid state: it has almost no viscosity and a high effective thermal conductivity. These properties are used to cool superconducting devices such as the Large Hadron Collider's magnets. From a theoretical point of view, superfluid dynamics can be described by a two-fluid model in which helium superfluid (He II) is seen as a mixture of fluid particles that behave as a classic fluid, and of superfluid particles having zero viscosity. When the temperature decreases, the volume fraction of superfluid increases, and the volume fraction of classic fluid decreases. Therefore, at very low temperatures drag forces due to viscous forces vanish, and classic concepts for describing flow and transport in porous media including permeability and tortuosity are challenged.

In collaboration with the Institut de Mécanique des Fluides de Toulouse (IMFT), the Commissariat à l'Energie Atomique (CEA), and the European Organization for Nuclear Research (CERN), I studied the theory of helium superfluid (He II) in porous media. To improve general knowledge on superfluid dynamics, I developed and verified a numerical tool called *HellFOAM*[5] that simulates He II at the pore-scale (Soulaine et al. 2015, 2017a). *HellFOAM*, released in the public domain, is the only simulator, so far, able to simulate superfluid helium solving the full two-fluid model. It is a powerful tool to support new experiments and to design enhanced cooling devices for superconducting magnets and quantum computers.

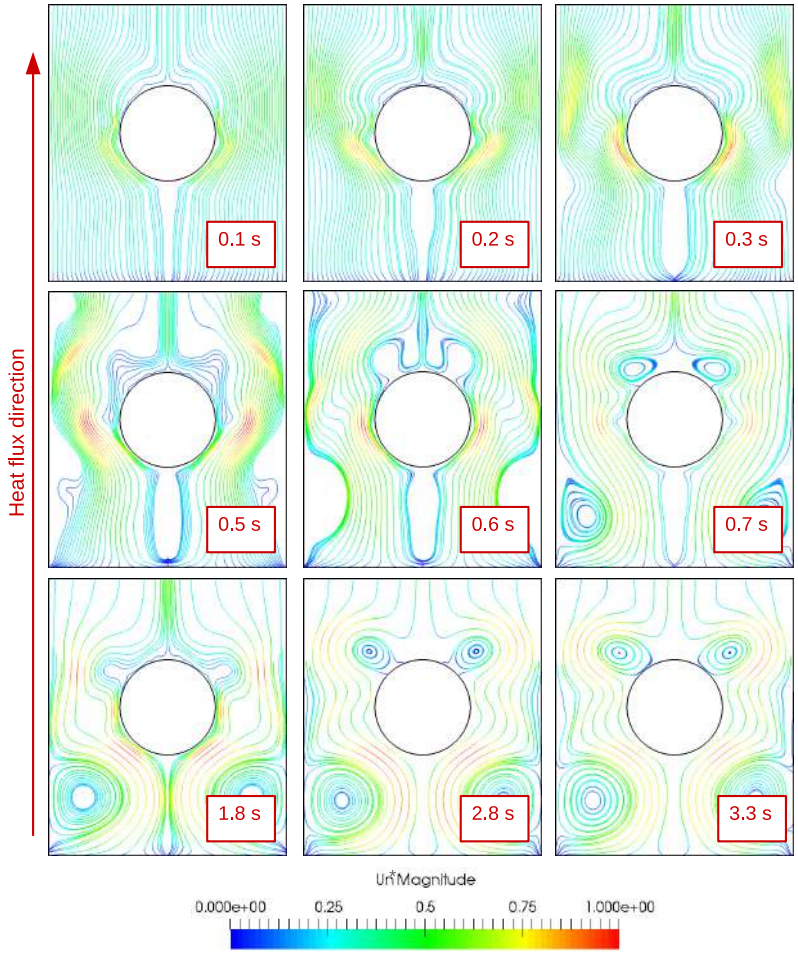


Figure 2.4: Plot of the streamlines of the classic fluid particles in flow of helium superfluid around a solid cylinder (from Soulaine et al. (2017a)).

Thanks to *HellFOAM*, we obtained two fundamental findings already. First, in Soulaine et al. (2017a), I was able to reproduce and explain, the unexpected observations about thermal counterflow of He II past a cylinder reported by Zhang and Van Sciver (2005) in Nature Physics using microfluidic experiments. Unlike classic fluids, two eddies are observed upstream the obstacle in addition to the two downstream eddies (see Fig. 2.4). The apparition of these two additional eddies is related to quantum turbulence that we analyzed by introducing a new dimensionless number. These results not only comfort the validity of the two-fluid model but they are also the first step toward a full investigation of superfluid flow in porous media

[5] https://github.com/csoulain/HellFOAM

that are conceptualized as an ensemble of solid cylinders. Then, I also showed that the temperature increase in a forced flow of He II was not only due to the Joules-Thomson effect as commonly believed but that quantum turbulence also generates a temperature elevation of the same order of magnitude (Soulaine et al. 2014b). Using this additional thermal effect, we were able to reproduce with high accuracy the experimental temperature profiles of Fuzier and Van Sciver (2008).

2.2.5 Development of column/reservoir-scale simulators

Since 2008, I have been involved in the development of several simulators for flow and transport in porous media to model processes at the scale of a column or a reservoir. Before my Ph.D., at Air Liquide Research Center, I developed coupled heat and mass transfer models to simulate gas separation processes in columns containing adsorbent nanoporous beads including silica gel, activated carbon, and zeolite (Soulaine 2008). The two-phase Darcy solver I developed during my Ph.D. has been improved, extended, and released under open-source licenses (Horgue et al. 2015). I was also involved in the development of *richardsFoam*, an open-source platform for simulating transport processes in the unsaturated zone using Richards equation (Orgogozo et al. 2014).

At ISTO, I am the lead architect of *porousMedia4Foam*, a comprehensive package for solving flow, transport, and geochemical processes within the OpenFOAM open-source platform (Soulaine et al. 2021c). Unlike all the other porous media simulators, *porousMedia4Foam* relies on micro-continuum models – a concept that I am developing continuously since 2013 (Carrillo et al. 2020; Soulaine et al. 2019, 2016, 2021c, 2017b, 2018; Soulaine and Tchelepi 2016)– and can solve pore-scale and continuum-scale problems as well as a combination of both scales. An overview of the micro-continuum approaches is given in Chapter 6. An in-depth introduction to the *porousMedia4Foam* is given in Chapter 7.

2.3 Curriculum vitae and list of publications

(last update: 27 Oct, 2021)

CYPRIEN SOULAINE

Associate Scientist at CNRS

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Github *Linkedin* *Skype*

Education

2009–2012 **Ph.D.**, *Institute of Fluid Mechanics, INP-Toulouse*[1], France.
Supervisor: Prof. Michel Quintard

2005–2008 **Master in Mechanical Engineering**, *MATMECA, INP-Bordeaux*[2], France.

Current position

2019–now **Associate (tenured) Scientist**, *CNRS*[3], Orléans, France.
Earth Sciences Institute of Orleans (ISTO) – UMR 7327, University of Orléans

Previous positions

2018–2019 **Research Scientist**, *BRGM*[4] *– French Geological Survey*, Orléans, France.
Department of Water, Environment and Ecotechnologies (in Dr. Francis Claret's group)
Multi-scale reactive transport modelling in geological formations.

2015–2018 **Research Associate**, *Stanford University*, California, USA.
Department of Energy Resources Engineering (in Prof. Hamdi Tchelepi's group)
Multi-scale modelling of multiphase flow and transport in porous media.

2013–2015 **Post-doctoral Researcher**, *Stanford University*, California, USA.
Department of Energy Resources Engineering (in Prof. Hamdi Tchelepi's group)
Pore-scale modelling for complex geological formations.

2012–2013 **Post-doctoral Researcher**, *Fluid Mechanic Institute of Toulouse, INP-Toulouse*, France.
in collaboration with CERN, the European Organization for Nuclear Research
Superfluid helium flow in porous media.

2009–2012 **PhD fellow**, *Fluid Mechanic Institute of Toulouse*, France.
Multi-scale modelling of cryogenic air distillation processes.

2008–2009 **R&D Engineer**, *Air Liquide Research Center*, Paris area, France.
Adsorption processes for hydrogen production plants.

Fellowships & Awards

2020 **Award of the French Ministry of Higher Education, Research and Innovation** for managing research (PEDR, rate of success <10%)

2019 **Early-Career Scientist Grant** from the French National Research Agency (rate of success <12%)

2019 **Laureate** of the French National **CNRS competition** for an Associate Scientist position

2013 **Toulouse Academy of Science Award**, best thesis in Engineering

2012 **Léopold Escande Award** from INP-Toulouse for outstanding PhD thesis (top 10%)

2009 PhD fellowship from Air Liquide (CIFRE Industrial and Innovation fellowship)

[1] INP-Toulouse: National Polytechnic Institute of Toulouse
[2] INP-Bordeaux: National Polytechnic Institute of Bordeaux
[3] CNRS: French National Centre for Scientific Research
[4] BRGM: French Geological Survey

Teaching activities

2021 – now Fundamental of Fluid Mechanics, lectures (10h), Polytech Orléans, France

2020 – now Fundamental of Porous Media Fluid Mechanics, lectures (20h), University of Pau, France

2019 – now Transport Phenomena in Porous Media, lectures (12h), University of Orléans, France

2014– 2018 Pore-scale simulation and modelling, lectures (3h), Stanford University

2014 Computational Fluid Dynamics, tutorials (18h), Stanford University

2012 Computational Fluid Dynamics, lectures & tutorials (24h), INP-Toulouse, France

Supervision of graduate students

2021 – now Dr. Flore Rembert (♀): Postdoc

2021 – now Laurez Maya (♂): PhD student

2020 – now Mojtaba Nourouzisadeh (♂): PhD student

2019 – now Dr. S. Pavuluri (♂): Postdoc

2018 – now co-mentoring of F. Carrillo (♂) PhD student at Princeton University with Prof Ian Bourg

2009 – now 1 to 3 undergraduate students per year for short term projects (1 to 6 months)

Previous

Period	Name		Diploma	Next position
2021	I. Belaid	♀	MSc Univ Gustave Eiffel	–
2020-2021	L. Maya	♂	MSc Polytech Nancy	PhD Candidate Univ Orléans
2014-2018	M. Abu AlSaud	♂	PhD Stanford Univ	Research Scientist at Saudi Aramco
2014-2016	M. Graveleau	♀	MSc Stanford Univ	Research Scientist at Lyft
2013-2015	A. AlZayer	♂	MSc Stanford Univ	Engineer at Saudi Aramco
2014-2016	M. Crémon	♂	MSc Stanford Univ	PhD Candidate Stanford Univ
2013-2016	Y. Wang	♂	MSc Stanford Univ	PhD Candidate Stanford Univ
2013	F. Houdroge	♀	MSc Toulouse Univ	PhD Candidate Monash Univ

Organization of scientific meetings

2021–2024 Organization committee of 6th Cargèse's summer school on flow and transport in porous media (July 2024). About 100 participants.

2010 – now **Organizer and instructor of short courses** (6 to 18h of lectures & tutorials) on Computational Fluid Dynamics in Porous Media with OpenFOAM[5] taught in many Universities (Stanford University, Beijing Normal University, University of Orléans, Geosciences Montpellier, CNRS), industries (Air Liquide, BRGM), and in international workshops (9th & 11th Interpore, 4th & 5th Cargèse's summer school on flow and transport in porous and fractured media). Each events involve 12 to 24 participants. **Overall I have trained more than 300 people.**

2020 – now Organization of FOAM-U (French and Belgian Users of OpenFOAM) Annual meeting (60 to 100 attendees)

2016 – now Session convener at Interpore's International Conference on Porous Media

2016 Session convener at XXII International Conference Computational Methods in Water Resources

2015 Session convener at SIAM Geosciences meeting.

Institutional responsabilities and reviewing activities

2020 – now **Head of the Porous Media Research Group** at ISTO

2019 – now **Reviewer for research proposals**: Swiss National Supercomputing Center (CSCS), American Chemical Society Petroleum Research Fund (ACS PRF)

[5]OpenFOAM is an open-source simulation platform

2011 – now **Referee for peer-reviewed journals**: Journal of Fluid Mechanics, Transport in Porous Media, Advances in Water Resources, Journal of Hydrology, Water Resources Research, Oil & Gas Science and Technology, Geophysical Research Letters, Chemical Engineering Sciences, Journal of Colloid and Interface Science, Scientific Report, Computer & Fluids, Numerical and Analytical Methods in Geomechanics

2013 – 2018 Organization of weekly seminars on pore-scale modelling and experiments, Stanford University

Memberships of scientific societies

2011 – now Member of Interpore, International Society for Porous Media

2014 – now Member (organizing committee) of FOAM-U, French and Belgian Users of OpenFOAM

Past and current notable collaborations

- **Prof. Hamdi Tchelepi** (multiscale modelling) & **Prof. Anthony Kovscek** (microfluidic experiments), Department of Energy Resources Engineering, Stanford University, CA, USA
- **Dr. Julien Maes** (numerical modelling of multiphase flows), Institute of GeoEnergy Engineering, Heriot-Watt University, U.K.
- **Prof. Ian Bourg** (molecular dynamics), Department of Civil and Environmental Engineering, Princeton University, NJ, USA
- **Ass. Prof. Catherine Noiriel**, (X-ray micro-tomography), Géosciences Environnement Toulouse, University of Toulouse, France
- **Ass. Prof. Sophie Roman**, (microfluidics), Earth Sciences Institute of Orléans, University of Orléans, France

PhD and MSc defense committee

2021 Evaluator of PhD thesis Yixun Sun, Polytechnique Montréal, Canada

2021 Evaluator of MSc thesis Laurez Maya, Polytech Nancy, France

2021 Evaluator of MSc thesis Viktor Gredicak, University of Zagreb, Croatia

2020 Evaluator of PhD thesis Valentin Jules, IPGP / Sorbonnes University, France

Career breaks

2015 Paternity leave of 1.5 months

Visiting periods

2019 Princeton University, Civil and Environmental Engineering. Invited by Prof. Ian Bourg during summer 2019.

Participation to research projects

2022–2026 **ANR IMAGE**, *Work Package Leader*, Induced polarization for exploring organic pollutant transport and bioremediation geoprocesses.
Multi-scale reactive transport and induded polarization modelling from nano to field scale.

2022–2026 **ANR JCJC GeoMIME**, Microfluidics for controlling multiphase and species transport towards clean energy systems in the subsurface.

2021 **CNRS MITI EsCapaDeS**, *co-PI*, Estimation of CO_2 storage capacity in geological formations.
Investigation of capillary trapping mechanisms using computational and experimental microfluidics

2019–2023 **ANR JCJC FraMatI**, *PI*[6], Microscale modeling of fracture-matrix interactions.
Experimental and computational modelling of reactive transport in unsaturated fractured porous media

[6] PI = Principal Investigator

2020–2021 **CNRS MITI CaraMBar**, *PI*, Characterization and Modeling of Geological Barriers.
Investigation of confinement barriers using computational and experimental microfluidics

2020–2021 **Labex Voltaire TransCol**, *PI*, Transport of colloids in porous media.
Experimental and computational modelling of colloidal transport in porous media

2019–2020 **ANCRE PORE-REACTIF**, *PI*, Modelling of hydro-geochemical processes.
Modelling of coupled flow and transport with geochemical packages

2019 **BRGM TRIPHASIQUE**, *PI*, Three-phase flow in soils and the subsurface.
Towards predictive modelling capabilities for three-phase flow in porous media

2018–2022 **ANR CATCH**, Multi-scale modelling of structure–chemistry–transport couplings.
Pore-scale modelling of reactive transport in geological formations.

2014–2018 **DOE–EFRC–NCGC**, Center for nanoscale controls on geologic CO_2.
Multi-scale two-phase reactive transport modelling in geological formations.

2014–2018 **TOTAL STEMS**, Stanford-Total Enhanced Modeling of Source Rocks.
Multi-scale analysis of source rocks

2012–2013 **IMFT-CERN-CEA**, Superfluid helium flow in porous media.
Development of numerical tools to simulate superfluid flow in porous media

2009–2012 **Air Liquide**, Cryogenic distillation columns.
Multi-scale analysis of processes occuring in distillation columns equipped with structured packing

On-going grants as Principal Investigator

Title	*Funding Source*	*Amount (k€)*	*Period*	*Short description*
FraMatI	ANR [7]	220	2019-2024	**Early-career Scientist Grant**. Microscale modeling of fracture-matrix interactions under reactive two-phase flow.
PORE-REACTIF	ANCRE[8]	140	2019-2020	Fellowship for the 12-months of Dr. Saideep Pavuluri post-doc on coupling flow and transport with geochemical packages at the continuum-scale.
TransCol	LabEx Voltaire	25	2020-2021	Experimental and computational modelling of colloidal transport in porous media.
CaraMBar	CNRS MITI	25	2020-2021	Investigation of confinement barriers using computational and experimental microfluidics.
ConvReact	CaSciModOT[9]	5	2021	Fellowship for 6-months for Imène Belaid master internship on modeling reactive convection in hydrothermal systems.

[9]French National Agency for Research
[9]Alliance Nationale de Coordination de la Recherche pour l'Energie
[9]Calcul Scientifique et Modélisation Orléans Tours

Publications

26 articles published in peer-reviewed international journals (14 as first authors)

3 articles in proceedings of international conferences (2 as first author)

31 oral presentations at international conferences

h-index: 16 (Google Scholar); 14 (Scopus)

52 co-authors

Preprints and articles under review

S. Pavuluri, C. Tournassat, F. Claret, **C. Soulaine**. Reactive transport modelling with a coupled OpenFOAM®-PHREEQC platform. *under review*

F. Carrillo, **C. Soulaine**, I. C. Bourg. The Impact of Sub-Resolution Porosity on Numerical Simulations of Multiphase Flow. *arXiv e-prints*, arXiv:2109.13903

Moataz O. Abu AlSaud, **Cyprien Soulaine**, Amir Riaz, Hamdi A. Tchelepi. Level-set method for accurate modeling of two-phase immiscible flow with moving contact lines. *arXiv e-prints*, arXiv:1708.04771

Articles published in peer review journals

2021 **Cyprien Soulaine**, Saideep Pavuluri, Francis Claret, and Christophe Tournassat. *porousMedia4Foam*: Multi-scale open-source platform for hydro-geochemical simulations with openfoam. *Environmental Modelling and Software*, volume 145, page 105199, 2021.

2021 **Cyprien Soulaine**, Julien Maes, and Sophie Roman. Computational microfluidics for geosciences. *Frontiers in Water*, volume 3, pages 1–11, 2021.

2021 **Cyprien Soulaine**, Laurence Girolami, Laurent Arbaret, and Sophie Roman. Digital rock physics: computation of hydrodynamic dispersion. *Oil & Gas Science and Technology – Rev. IFP Energies nouvelles*, volume 76, 2021.

2021 Catherine Noiriel and **Cyprien Soulaine**. Pore-scale imaging and modelling of reactive flow in evolving porous media: tracking the dynamics of the fluid-rock interface. *Transport in Porous Media*, volume 140, pages 181—213, 2021.

2020 Sophie Roman, **Cyprien Soulaine**, and Anthony Kovscek. Pore-scale visualization and characterization of viscous dissipation in porous media. *Journal of Colloid and Interface Science*, volume 558, pages 269–279, 2020.

2020 J. Poonoosamy, **C. Soulaine**, A. Burmeister, G. Deissmann, D. Bosbach, and S. Roman. Microfluidic flow-through reactor and 3d raman imaging for in situ assessment of mineral reactivity in porous and fractured porous media. *Lab-on-a-Chip*, 2020.

2020 S Molins, **C Soulaine**, NI Prasianakis, A Abbasi, Philippe Poncet, AJC Ladd, V Starchenko, S Roman, D Trebotich, HA Tchelepi, and CI Steefel. Simulation of mineral dissolution at the pore scale with evolving fluid-solid interfaces: Review of approaches and benchmark problem set. *Computational Geosciences*, pages 1–34, 2020.

2020 Julien Maes and **Cyprien Soulaine**. A unified single-field volume-of-fluid-based formulation for multi-component interfacial transfer with local volume changes. *Journal of Computational Physics*, volume 402, page 109024, 2020.

2020 Francisco J. Carrillo, Ian C. Bourg, and Cyprien Soulaine. Multiphase flow modeling in multiscale porous media: An open-source micro-continuum approach. *Journal of Computational Physics: X*, volume 8, page 100073, 2020.

2019 **Cyprien Soulaine**, Patrice Creux, and Hamdi A. Tchelepi. Micro-continuum framework for pore-scale multiphase fluid transport in shale formations. *Transport in Porous Media*, 2019.

2018 **Cyprien Soulaine**, Sophie Roman, Anthony Kovscek, and Hamdi A Tchelepi. Pore-scale modelling of multiphase reactive flow. Application to mineral dissolution with production of CO_2. *Journal of Fluid Mechanics*, volume 855, page 616–645. Cambridge University Press, 2018.

2018 Julien Maes and **Cyprien Soulaine**. A new compressive scheme to simulate species transfer across fluid interfaces using the volume-of-fluid method. *Chemical Engineering Science*, volume 190, pages 405–418. Elsevier, 2018.

2017 **Cyprien Soulaine**, Sophie Roman, Anthony Kovscek, and Hamdi A Tchelepi. Mineral dissolution and wormholing from a pore-scale perspective. *Journal of Fluid Mechanics*, volume 827, pages 457–483. Cambridge University Press, 2017.

2017 **Cyprien Soulaine**, Michel Quintard, Bertrand Baudouy, and Rob Van Weelderen. Numerical investigation of thermal counterflow of he-ii past cylinders. *Physical Review Letters*, volume 118, page 074506, February 2017.

2017 Marguerite Graveleau, **Cyprien Soulaine**, and Hamdi A Tchelepi. Pore-scale simulation of interphase multicomponent mass transfer for subsurface flow. *Transport in Porous Media*, volume 120, pages 287–308. Springer, 2017.

2016 **Cyprien Soulaine** and Hamdi A. Tchelepi. Micro-continuum approach for pore-scale simulation of subsurface processes. *Transport In Porous Media*, volume 113, pages 431–456, 2016.

2016 **Cyprien Soulaine**, Filip Gjetvaj, Charlotte Garing, Sophie Roman, Anna Russian, Philippe Gouze, and Hamdi Tchelepi. The impact of sub-resolution porosity of x-ray microtomography images on the permeability. *Transport in Porous Media*, volume 113, pages 227–243, May 2016.

2016 Sophie Roman, **Cyprien Soulaine**, Moataz Abu AlSaud, Anthony Kovscek, and Hamdi Tchelepi. Particle velocimetry analysis of immiscible two-phase flow in micromodels. *Advances in Water Resources*, volume 95, pages 199–211, September 2016.

2015 **Cyprien Soulaine**, Michel Quintard, Hervé Allain, Bertrand Baudouy, and Rob Van Weelderen. A PISO-like algorithm to simulate superfluid helium flow with the two-fluid model. *Computer Physics Communications*, volume 187, pages 20–28, 2015.

2015 Pierre Horgue, **Cyprien Soulaine**, Jacques Franc, Romain Guibert, and Gérald Debenest. An open-source toolbox for multiphase flow in porous media. *Computer Physics Communications*, volume 187, pages 217–226. Elsevier, 2015.

2014 **Cyprien Soulaine** and Michel Quintard. On the use of a Darcy–Forchheimer like model for a macro-scale description of turbulence in porous media and its application to structured packings. *International Journal of Heat and Mass Transfer*, volume 74, pages 88 – 100, 2014.

2014 **Cyprien Soulaine**, Pierre Horgue, Jacques Franc, and Michel Quintard. Gas-liquid flow modeling in columns equipped with structured packing. *AIChE Journal*, volume 60, pages 3665–3674, 2014.

2014 L. Orgogozo, N. Renon, **C. Soulaine**, F. Hénon, S.K. Tomer, D. Labat, O.S. Pokrovsky, M. Sekhar, R. Ababou, and M. Quintard. An open source massively parallel solver for richards equation: Mechanistic modelling of water fluxes at the watershed scale. *Computer Physics Communications*, volume 185, pages 3358 – 3371, 2014.

2013 **Cyprien Soulaine**, Yohan Davit, and Michel Quintard. A two-pressure model for slightly compressible single phase flow in bi-structured porous media. *Chemical Engineering Science*, volume 96, pages 55 – 70, 2013.

2013 Hervé Allain, Rob Van Weelderen, Bertrand Baudouy, Michel Quintard, Marc Prat, and **Cyprien Soulaine**. Investigation of suitability of the method of volume averaging for the study of heat transfer in superconducting accelerator magnet cooled by superfluid helium. *Cryogenics*, volume 53, pages 128–134. Elsevier, 2013.

2011 **Cyprien Soulaine**, Gérald Debenest, and Michel Quintard. Upscaling multi-component two-phase flow in porous media with partitioning coefficient. *Chemical Engineering Science*, volume 66, pages 6180–6192, 2011.

Conference proceedings published in journals with peer review

C. Soulaine, M. Quintard, H. Allain, B. Baudouy, R. Van Weelderen, « Numerical Investigation of Heat Transfer In a Forced Flow of He II » *Proceedings of the 15th International Heat Transfer Conference*, IHTC-15 August 10-15, 2014, Kyoto, Japan, 2014

Conference proceedings published in journals

J.Maes and **C. Soulaine**, «Direct Pore-Scale Modelling Of Dissolution And Trapping Of Supercritical CO_2 In Reservoir Brine » *Proceedings of the 16th European Conference on the Mathematics of Oil Recovery*, ECMOR XVI, 3-6 September 2018, Barcelona, Spain

C. Soulaine and H. A. Tchelepi, « Micro-continuum Formulation for Modelling Dissolution in Natural Porous Media » *Proceedings of the 15th European Conference on the Mathematics of Oil Recovery*, ECMOR XV, 29 August – 1 September 2016, Amsterdam, Netherlands

Oral communications

Presentation in conferences and workshops

2021 "Computational microfluidics for geosciences - Simulation techniques to model flow and transport at the pore-scale", 5th Cargèse summer school on flow and transport in porous and fractured media, July 20–31, 2018. Cargèse, France ***(invited speaker)***

2021 « A Micro-Continuum Approach for Solving Two-Phase Flow in Porous Media at Different Scales», SIAM Conference on Mathematical and Computational Issues in the Geosciences, June 21–24, 2021. Online Conference. ***(invited speaker)***

2020 « A (real) multi-scale solver for two-phase flow: a micro-continuum approach», InterPore 2020, 12th Annual Meeting, Aug 31 – Sept 4, 2020. Virtual Conference

2019 « Introduction to open-source simulation using OpenFOAM», 31ème journée de la fédération de Calcul Scientifique et Modélisation à Orléans et Tours, December 17, 2019. Orléans, France ***(invited speaker)***

2019 « Pore-scale modelling of multiphase reactive flow», SITRAM19 - Workshop Advances in the SImulation of reactive flow and TRAnsport in porous Media, December 2–3, 2019. University of Pau, France

2019 « Multi-scale modeling of acid stimulation processes in carbonate formations», DEEPEGS international symposium, November 6–8, 2019. Orléans, France

2019 « Quantifying the reactive surface area from pore-scale simulations », Le Studium Workshop - Knowledge's frontiers in water unsaturated hydrogeosystems: interface dynamics, heterogeneities & couplings, June 27–28, 2019. Orléans, France

2019 « OpenFOAM and Geosciences», 4th French/Belgian OpenFOAM® users conference, June 12–13, 2019. Marseille, France

2019 "Pore-scale modeling of coupled hydro-geological processes", InterPore 11th Annual Meeting, May 6–10, 2019. Valencia, Spain

2019 « Superfluid and quantum turbulence in porous media», Workshop upscaling in porous media: A tribute to Michel Quintard, April 15–16, 2019. I2M Bordeaux, France ***(invited speaker)***

2018 « Digital Rock Physics: Objectives and Challenges», 29ème journée de la fédération de Calcul Scientifique et la Modélisation à Orléans et Tours, December 12, 2018. Orléans, France ***(invited speaker)***

2018 "Micro-Continuum modeling: an hybrid-scale approach", Gordon Research Conference on Flow & Transport in Permeable Media, July 9–12, 2018. Sunday River, Newry, ME, USA ***(invited speaker)***

2018 "Simulation techniques to model flow and transport at the pore-scale", 4th Cargèse summer school on flow and transport in porous and fractured media, June 25 – July 7, 2018. Cargèse, France ***(invited speaker)***

2018 "Superfluid and quantum turbulence in porous media", InterPore 10th Annual Meeting and Jubilee, May 14–17, 2018. New Orleans, USA

2017 "Micro-Continuum Approach for Pore-Scale Simulation of Subsurface Processes", MATHIAS Conference, Oct 25–27, 2017. Marne-la-Vallée, France

2017 "Pore-Scale Simulation of Reactive Mass Transport with a Micro-Continuum Approach", Reactive Transport Workshop, Oct 2–5, 2017. Amboise, France

2017 "Micro-continuum approach for pore-scale simulation of subsurface processes", SIAM Conference on Mathematical and Computational Issues in the Geosciences, Sept 11–14, 2017. Erlangen, Germany

2017 "Micro-Continuum Approach for Pore-Scale Simulation in Subsurface Processes", TOTAL Pore-Scale Workshop, Pau, France. June 8–9, 2017

2017 "Mineral dissolution and wormholing from a pore-scale perspective", InterPore 9th International Conference on Porous Media & Annual Meeting, May 8–11, 2017. Rotterdam, Netherlands

2016 "Micro-continuum approach for pore-scale simulation of subsurface processes", 53rd Annual Technical Meeting of the Society of Engineering Science, Oct 2–5, 2016. University of Maryland, College Park, USA ***(invited speaker)***

2016 "Micro-continuum Formulation for Modelling Dissolution in Natural Porous Media", 15th European Conference on the Mathematics of Oil Recovery, ECMOR XV, Aug 29 – Sept 1, 2016. Amsterdam, Netherlands

2016 "Pore-Scale Simulation of sCO_2/Brine Dissolution", XXII International Conference Computational Methods in Water Resources, CMWR 2016, June 20–24, 2016. Toronto, Canada

2016 "A Multiphysics Multiscale Framework for Modeling Mass and Heat Flow at Small Scales: Carbonate Dissolution", XXII International Conference Computational Methods in Water Resources, CMWR 2016, June 20–24, 2016. Toronto, Canada

2015 "Multi-scale simulation for flow, heat and mass transport in porous media based on a Darcy-Brinkman model", MATHIAS Conference, Oct 28–30, 2015. Paris, France

2015 "Pore-scale Simulation of Carbonate Dissolution", SIAM Conference on Mathematical and Computational Issues in Geosciences, June 29 – July 5, 2015. Stanford, CA, USA

2015 "Characterization of Type I Source-Rock by Pore-scale Simulation", Engineering Mechanics Institute Conference, EMI 2015, June 16–19, 2015, Stanford, CA, USA

2015 "Investigation of Two-Phase Flow Mechanisms in Porous Media Using Micro-Particle Image Velocimetry", InterPore 7th International Conference on Porous Media & Annual Meeting. May 18–21, 2015, Padova, Italy

2014 "Simulation of multiphase flow at the pore scale: doable, useful?", AGU Fall Meeting 2014, San Francisco, USA ***(invited speaker)***

2014 "Numerical Investigation of Heat Transfer In a Forced Flow of He II" 15th International Heat Transfer Conference, IHTC-15 August 10–15, 2014, Kyoto, Japan

2013 "Gas-Liquid flow modeling in columns equipped with structured packing seen as bi-structured porous media", AIChE Annual Meeting 2013. San Francisco, CA, USA

2012 "Two-phase flow solver for anisotropic porous media", 7th OpenFOAM Workshop, June 25-28, 2012, Darmstadt, Germany

2012 "Derivation of an anisotropic Darcy-Forchheimer equation including turbulence effects and its application to structured packings", Interpore 4th International Conference on Porous Media & Annual Meeting of the International Society for Porous Media, May 14–16, 2012. Purdue University, West Lafayette, Indiana, USA

2010 "Upscaling Multicomponent Two-Phase Dispersion with Partitioning Coefficient", AIChE Annual Meeting 2010, Nov 7–12, 2010. Salt Lake City, Utah, USA

2010 "Changement d'échelle d'un écoulement diphasique multiconstituants avec relation de partitionnement", 10ème édition des Journées d'Etude sur les Milieux Poreux, Oct 20–21, 2010. Nancy, France

Poster

2018 "Multiphase micro-continuum models: a hybrid-scale approach", InterPore 10th Annual Meeting and Jubilee, May 14-17, 2018. New Orleans, USA

2017 "Pore-Scale Simulation of the Processes Associated with the Injection and Sequestration of CO2 in the Subsurface", 2017 EFRC-Hub-CMS PI Meeting, July 24-25, 2017. Washington, D.C., USA

2017 "Mineral dissolution and wormholing from a pore-scale perspective", InterPore 9th International Conference on Porous Media & Annual Meeting, May 8 - 11, 2017. Rotterdam, Netherlands

2015 "Characterization of type I source rock by pore-scale simulations", InterPore 7th International Conference on Porous Media & Annual Meeting. May 18 - 21, 2015, Padova, Italy

2014 "Pore-scale modeling of oil-shale pyrolysis with CFD-DEM method", Gordon Research Conference on Flow and Transport in Permeable Media, July 6 - 11, 2014. Bates College, Lewiston, ME, US

Seminar

2021 "Micro-continuum approach for the multi-scale modeling of coupled processes in the subsurface", Geological Research Seminars, Ghent University. Oct 22, 2021.

2021 "Multi-scale modelling with micro-continuum approach", MultiscaleModelling and Heterogeneous Media Research Theme Seminar, University of Nottingham. Apri 1, 2021.

2020 "Approches micro-continues pour la modélisation multi-échelle des écoulements diphasiques en milieux poreux", IFP Enérgie Nouvelle, France. December 10, 2021

2020 "Micro-continuum approach for multi-scale modelling of subsurface processes", Center of Excellence PoreLab, Department of Geoscience and Petroleum, Norwegian University of Science and Technology, Norway. December 2, 2020

2020 "Micro-continuum modelling: A hybrid-scale approach for solving coupled processes in porous media", University of Huddersfield, Huddersfield, UK. February 28, 2020.

2020 "Micro-continuum modelling: A hybrid-scale approach for solving coupled processes in porous media", University of Portsmouth, Portsmouth, UK. February 26, 2020.

2019 "Multi-scale modeling of coupled hydro-geochemical processes in porous and fractured media", Institut für Energie und Klimaforschung (IEK-6), Forschungszentrum Jülich, Jülich, Germany. November 28, 2019

2019 "Multi-scale modeling of coupled hydro-geochemical processes in porous and fractured media", Department of Mathematics, Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany. October 25, 2019

2019 "Multi-scale modeling of coupled hydro-geochemical processes in porous and fractured media", Department of Civil and Environmental Engineering, Princeton University, Princeton, NJ, USA. August 26, 2019

2019 "Modélisation à l'échelle des pores des processus hydro-géochimiques dans les réservoirs poreux et fracturés", Institut de Physique du Globe de Paris, Paris, Jan 15, 2019

2018 "Multi-Scale Analysis of Hydro-Geochemical Instabilities in Porous Media", Laboratoire Matière et Systèmes Complexes - Université Paris-Diderot, Paris, France. Dec 6, 2018

2018 "Pore-scale Modeling of Hydro-Geochemical Processes in Porous and Fractured Media", GéoRessources Nancy, Nancy, France. Nov 26, 2018

2017 "Mineral Dissolution and Wormholing from a Pore-Scale Perspective", Université de Pau et des Pays de l'Adour, Pau, France. June 6, 2017

2017 "Mineral Dissolution and Wormholing from a Pore-Scale Perspective", UC Berkeley, Mechanical Engineering, Berkeley, CA, USA. April 17, 2017

2016 "Pore-scale Modeling of Rock Dissolution", TU Delft - DARSim seminar, Delft, Netherlands. Sept 2, 2016

2016 "Pore-scale Modeling of Rock Dissolution", Heriot-Watt University, Edinburgh, Scotland. June 16, 2016

2015 "Simulation of flow in porous media with OpenFOAM®", Baker Hughes, Palo Alto, CA, USA. Jan 30, 2015

2013 "Modeling of Flows in Bi-Structured Porous Media", Energy Resources Engineering, Stanford University, Stanford, CA, USA. Oct 22, 2013

2013 "Modélisation des écoulements dans les garnissages structurés", Institut de Mécanique et d'Ingénierie de Bordeaux, Bordeaux, France. Feb 14, 2013

Chapter 3

On the origin of Darcy's law

When[1] *one thinks about porous media, the immediate concepts that come to mind are porosity, permeability, and Darcy's law. The latter is probably one of the most important tools in hydrodynamics and engineering presenting numerous applications in oil recovery, chemical engineering, nuclear safety, and many other engineering fields. It has been first formulated by the French engineer Henri Darcy in 1856 in Appendix D of his famous book* Fontaines publiques de la ville de Dijon *(Darcy 1856) to describe flow through beds of sand. His column experiments (see Figure 3.1) showed that the total discharge is proportional to the total pressure drop divided by the fluid viscosity, and that the linear coefficient, called permeability, is intrinsic to the investigated porous medium. Darcy's law is the equation that governs the flow of fluid in porous media. If one opens any fluid dynamics textbook, however, one learns that fluid motion is described by Navier-Stokes equations. So, what is this law? Are there any links between Darcy's law and fluid mechanics theory? This chapter introduces two different representations of the physics of fluid flow in porous media and their interrelation. These different approaches will lead us to the very definition of a porous medium.*

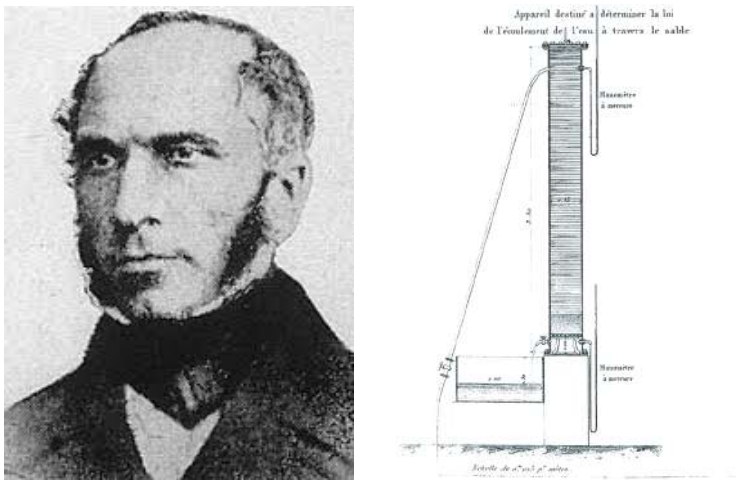


Figure 3.1: Henri Darcy and its famous sand column experiment

[1] This chapter is adapted from a text I wrote in 2015 when I was a Research Associate at Stanford University. The document has been uploaded on my personal webpage, got some citations, but has never been published.

3.1 Two representations of the physics of fluid flow in porous media

A porous medium is a material that contains void spaces occupied by one or more fluid phases (gas, water, oil, etc.) and a solid matrix. A two-dimensional (2D) cross-section through the three-dimensional (3D) image of a sandstone sample is presented in Figure 3.2. The solid grains appear in white and the void or pore spaces are black. A fluid, denoted β, occupies pore space and we define this region as V_β as opposed to V_σ, the region occupied by the solid phase, σ. Figure 3.2 clearly highlights a sharp delineation between the solid and the void space: the interfacial area, denoted $A_{\beta\sigma}$. A lot of physico-chemical phenomena, such as surface reaction, may occur at this interface.

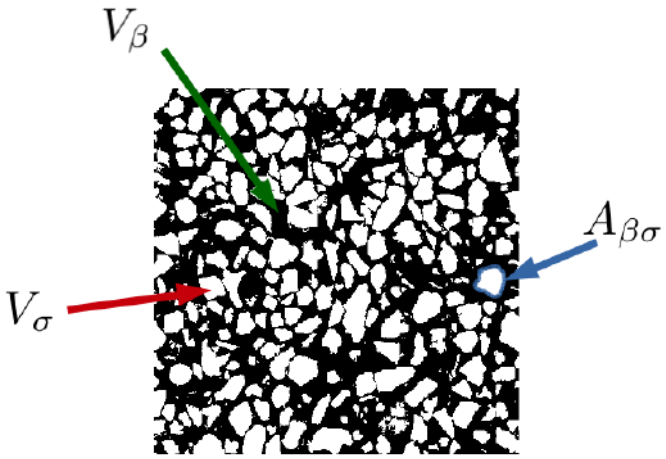


Figure 3.2: Two-dimensional slice of a sandstone. The black region represents the void space while the white region represents the solid grains.

Figure 3.2 also displays the high complexity of the pore network topology and morphology of real rocks. The local heterogeneities of the connected pore structure strongly influence the flow pathways. To characterize the flow in every location in V_β, we define a velocity, $\boldsymbol{v}_\beta$, and a pressure, p_β. Figure 3.3a, shows that a fluid flows through some preferential pathways leaving some area of the pore space stagnant. In natural porous media such as underground reservoirs, the flow is usually very slow. In that case, and assuming incompressible and Newtonian fluids, the flow patterns are independent of the flow rate and the type of the fluid. Only the magnitude of $\boldsymbol{v}_\beta$ changes linearly with the pressure drop. Actually, there are universal conservation laws that govern the fluid displacement in such conditions which are called the Stokes equations, a subset of the more general equations of fluid mechanics, the Navier-Stokes equations. For a given fluid of density ρ_β and viscosity μ_β, these equations read,

$$\begin{cases} \nabla.\boldsymbol{v}_\beta = 0 & \text{in } V_\beta \\ 0 = -\nabla p_\beta + \rho_\beta \boldsymbol{g} + \mu_\beta \nabla^2 \boldsymbol{v}_\beta & \text{in } V_\beta \end{cases} \tag{3.1}$$

For the time being, you can take these equations for granted. Further details and explanations are given in chapter 5. To be well-posed, this mathematical problem is completed with boundary conditions at the solid walls, $A_{\beta\sigma}$. For single-phase flow,

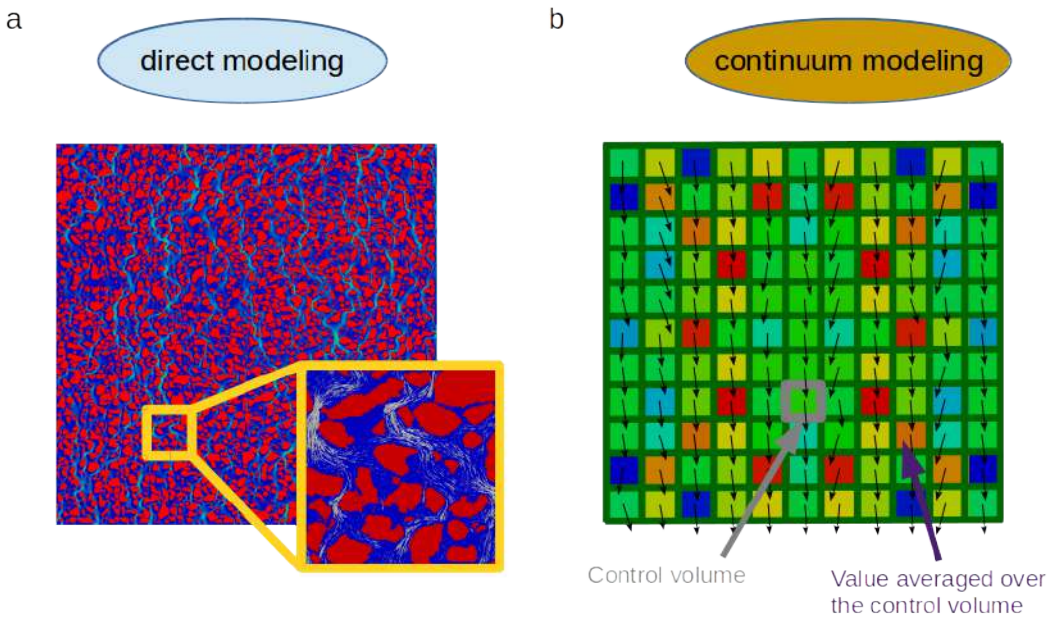


Figure 3.3: Two different representations of the physics of flow in porous media. The arrows represent the velocity vectors. (a) direct modelling or pore-scale approach where the solid is explicitly represented. (b) the continuum modelling or Darcy scale where the physics is governed by quantities averaged over control volumes. The color map represents the volume fraction of solid per control volume.

we consider no-slip conditions at the fluid/solid interface: for every point of $A_{\beta\sigma}$, $\boldsymbol{v}_\beta = 0$.

Solving the flow in a porous medium with Stokes equations is called *direct modelling* approach because the physics is directly solved without any assumptions in the microstructure geometry. The only input is the geometry of the solid skeleton and the fluid properties (viscosity and density). It is worth mentioning that for complex topologies Stokes equations do not have simple analytical solutions and need therefore to be approximated numerically. This approach is often referred to as *direct numerical simulation* (*DNS*) or *computational microfluidics*. Despite its accuracy and close representation of the physics, this approach has very severe limitations. Indeed, the geometry of the complex pore space enclosed by the solid skeleton must be described in complete detail. Although the last decade has seen tremendous progress in high resolution imaging techniques such as X-ray computed microtomography, this approach is not sustainable for large domains. It is of particular importance when investigating natural porous media. Even if you had the most powerful computer, how could you get the exact pore network topology of $100\,\mathrm{m}^3$ of rocks? As this representation of the physics of fluid flow in porous material often applies to small objects whose exact pore structure can be resolved, it is often referred to as *pore-scale modelling* or *pore-scale simulation*.

Another representation of the physics of fluid flow in a porous medium allows to deal with larger domains of investigation. In this representation, control volumes are defined all over the domain and quantities averaged over these volumes are considered (see Figure 3.3b). The control volume can not be taken arbitrarily since it needs to be a *Representative Elementary Volume (REV)* (Bear 1988). The very definition of the REV is often an integral part of the modelling challenge. Nevertheless,

assuming that the REV scale is known, we can define quantities averaged according to this control volume. With this approach, the pore space is no longer explicitly represented and we have instead information such as the volume fraction of void in a control volume. From now on, the fluid flow unknowns are the average velocity $\overline{\boldsymbol{v}}_\beta$ and the average pressure $\overline{p_\beta}^\beta$. These overline notations are used to emphasize the differences between this representation and the direct modelling approach. We will see in the next section how to compute these quantities. Because the pore network topology of the material is not explicit the fluid mechanics equations are no longer valid. In this representation, the averaged quantities are governed by Darcy's law,

$$\begin{cases} \nabla.\overline{\boldsymbol{v}}_\beta = 0, \\ \overline{\boldsymbol{v}}_\beta = -\frac{\mathsf{K}}{\mu_\beta}.\left(\nabla\overline{p_\beta}^\beta - \rho_\beta \boldsymbol{g}\right). \end{cases} \tag{3.2}$$

where the tensor K is the permeability, a constant intrinsic to the porous medium under investigation. It contains the information related to the pore space topology, or at least enough information to be representative of its geometry. Permeability is referred to as an *effective coefficient* because it translates information from a smaller scale. This approach is referred to as *continuum* or *Darcy modelling*. We also name it *macroscale* modelling as opposed to *microscale* modelling. Without going any further, we see that if one can get the pore space geometry of a REV and solves the flow in this REV, then by averaging the resulting velocity profile one can deduce the permeability.

Ideally, these two representations of the physics should produce the same results. They present, however, some fundamental differences in their formalism. The direct modelling has an explicit interface that delineates the solid and the void space where boundary conditions need to be specified and each point of the direct model domain contains either fluid or solid. Conversely, each point of the continuum-scale domain contains an aggregate of both fluid and solid. With such an approach, all the solid/fluid interfacial information is embedded in effective properties.

3.2 Volume averaging: from Stokes to Darcy

We have seen in the previous section that the physics of flow in porous media can be represented through two different approaches: direct and continuum modelling. The latter being an average version of the former. In this section, we provide a mathematical definition for the average quantities and we explain the origin of Darcy's law from the pore-scale physics.

Let's define a function, ζ_β, associated with the β-phase. It has value in the void space only. It can be a scalar, like for instance the pressure field, p_β or a vector, as the velocity field, $\boldsymbol{v}_\beta$. We introduce the average over the control volume, V, known as superficial average and defined as

$$\overline{\zeta}_\beta = \frac{1}{V}\int_{V_\beta} \zeta_\beta dV, \tag{3.3}$$

and the intrinsic phase average as

$$\overline{\zeta_\beta}^\beta = \frac{1}{V_\beta}\int_{V_\beta} \zeta_\beta dV. \tag{3.4}$$

For instance, the Darcy velocity, $\overline{v}_\beta$, is defined with this first average while the macroscale pressure, $\overline{p_\beta}^\beta$ is the average value in the void space only. Both operators are related by the trivial relation,

$$\overline{\zeta}_\beta = \phi \overline{\zeta_\beta}^\beta, \tag{3.5}$$

where $\phi \equiv \frac{V_\beta}{V}$ is the porosity, defined as the ratio of the volume occupied by the void (the pore volume) over the control volume (bulk volume). It measures the volume in a material that is occupied by fluids. In most cases the porous medium is assumed to be motionless and non-deformable, which means that ϕ does not vary over time and only depends on the solid geometry.

Now that the average operators are defined, and assuming that the REV exists, we can give a mathematical definition of porosity. It is based on the distribution of the void and solid that coexist in the control volume. Lets define the mask function, δ_β, in the entire domain V as,

$$\delta_\beta = \begin{cases} 1 & \text{in } V_\beta \\ 0 & \text{elsewhere} \end{cases}. \tag{3.6}$$

This function labels the void with 1 and the solid phase with 0. It can result from the process of segmentation of a raw image which allows to differentiating the solid from the void (see Figure 3.2). The integration of this function over a control volume, V, yields the volume, $V_\beta = \int_V \delta_\beta dV$, occupied by the void within V. The volume fraction of void or *porosity*, ϕ, is simply the superficial average of the indicator function[2],

$$\phi = \overline{\delta_\beta}. \tag{3.7}$$

Actually, we can think of $\overline{\cdot}$ as an average operator that is applied to Stokes equations to derive the governing equations for the average values. This upscaling process, however, is not straightforward. In particular, the average of a derivative is the derivative of the average plus the contribution of the boundary values at the solid walls. Mathematically it is performed with the following homogenization theorem,

$$\overline{\nabla \zeta_\beta} = \nabla \overline{\zeta_\beta} + \frac{1}{V} \int_{A_{\beta\sigma}} \boldsymbol{n}_{\beta\sigma} \zeta_\beta dA, \tag{3.8}$$

where $\boldsymbol{n}_{\beta\sigma}$ denotes the normal vector at the solid walls. The surface integral term in Eq. (3.8) translates the boundary value of the direct model to a volumetric source term in the Darcy-scale representation. This term is essential and has to be taken into account.

It took a long time to relate Darcy's law to the equations of fluid mechanics. This has been performed over the past decades by integrating the Stokes equations over a representative volume element of the porous medium (see for instance the work of Whitaker (1986a) who derived Darcy's law from the volume averaging of Stokes). These major breakthroughs in theoretical fluid mechanics brought new perspectives to Darcy's law. Actually, when following the theoretical upscaling from Stokes momentum equation, the resulting equation is not Darcy, but a more

[2]Note that by definition the intrinsic volume average of δ_β is always $\overline{\delta_\beta}^\beta = 1$.

comprehensive conservation law first proposed by Brinkman (1947) and referred to as Darcy-Brinkman or Darcy-Brinkman-Stokes equation. It reads,

$$0 = -\nabla \overline{p_\beta}^\beta + \rho_\beta \boldsymbol{g} + \mu_\beta^* \nabla^2 \overline{\boldsymbol{v}}_\beta - \mu_\beta \mathsf{K}^{-1}.\overline{\boldsymbol{v}}_\beta. \tag{3.9}$$

One recognizes the Stokes equation for the average velocity and pressure with an additional source term, $\mu_\beta \mathsf{K}^{-1}.\overline{\boldsymbol{v}}_\beta$. From a fluid mechanics point of view, this term is a drag force due to the viscous friction of the fluid with the walls of the solid structure. It comes from the surface integral in Eq. (3.8). It is the same force that gives a cyclist hard time when he bikes against the wind (see Figure 3.4). Therefore, the permeability is seen as a drag force coefficient. Compared to a bicycle, the solid surface per volume in a porous medium is very large, and so are the friction forces. The larger is the solid surface, the more important is the drag force. In practice, it turns out that most of the time, the viscous term, $\mu_\beta^* \nabla^2 \overline{\boldsymbol{v}}_\beta$, is negligible compared with $\mu_\beta \mathsf{K}^{-1}.\overline{\boldsymbol{v}}_\beta$, which leads then to Darcy's law. We will see in Chapters 5 and 6 situations where it is relevant to use Darcy-Brinkman equation.

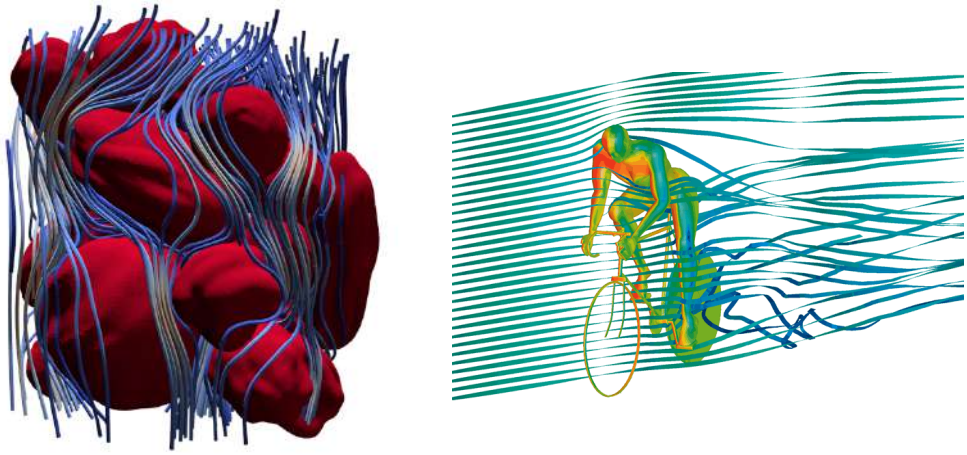


Figure 3.4: Streamlines through a porous medium (credit: F. Hénon) and a cyclist. In both cases, the friction of the fluid against the solid surfaces yields in drag forces.

Perhaps even more important than just the mathematical demonstration, the homogenization works that have lead to Darcy's equation emphasize that this law which was initially formulated as a phenomenological law for natural porous media with small pore throats can be extended to other porous media. All the media depicted in Figure 3.5 can be considered as porous media, with characteristic pore throat sizes ranging from few microns to few meters. For instance, the structured packings that equip air distillation columns for air separation or CO_2 capture are made of the same elementary pattern with pores of about a centimeter that are repeated millions of time within the column. The highest columns can reach up to 60 m high. It is clear that a pore-scale representation is not sustainable to simulate the flow in the entire column and that an averaged approach based on Darcy's law is more relevant (Soulaine et al. 2014a). For the same reason when investigating forest fires at large scale, it is not conceivable to represent all the branches and leaves, but instead, the canopy is considered as a porous medium where Darcy's law is valid.

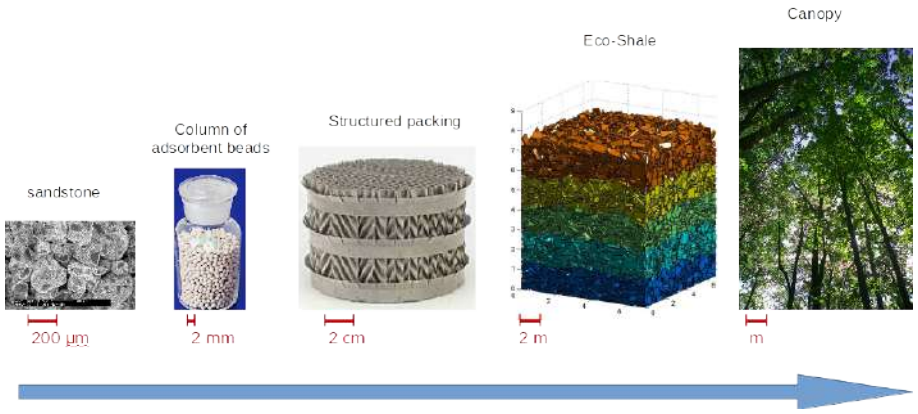


Figure 3.5: Example of porous media for a wide range of length scales.

3.3 Direct applications and analytical solutions

In few particular configurations, the Stokes equations have simple analytical solutions as, for instance, in the case of viscous flow between two parallel plates or flow through a capillary tube.

3.3.1 Viscous flow between parallel plates

In the case of viscous flow between two parallel plates (see Figure 3.6), we can suppose that there is a uniform effective pressure gradient in the x-direction, so that $\nabla p = \frac{\Delta P}{L}\boldsymbol{e}_x$. Moreover, considering that the dimensions of the plates are much

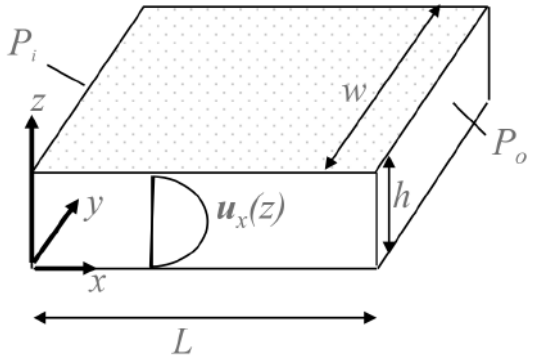


Figure 3.6: Schematic of flow between two parallel plates

larger than the thickness h, then we can consider that the flow is only carried by the x component and only depends on z, *i.e.*, $\boldsymbol{v}_\beta = v_x(z)\boldsymbol{e}_x$. Hence, the momentum equation reads

$$\mu_\beta \frac{\partial^2 v_x}{\partial z^2} = \frac{\Delta P}{L}, \tag{3.10}$$

with the non-slip boundary condition at the walls,

$$v_x\left(z = \pm\frac{h}{2}\right) = 0. \tag{3.11}$$

The integration of this boundary value problem leads to the following parabolic profile,

$$v_x(z) = \frac{1}{\mu_\beta}\frac{h^2}{8}\frac{\Delta P}{L}\left[1-\left(\frac{2z}{h}\right)^2\right]. \tag{3.12}$$

This is the 2D solution of a Poiseuille flow. We notice that the maximum velocity, $v_{z,max} = \frac{h^2}{8\mu_\beta}\frac{\Delta P}{L}$, is reached in the middle of the channel. Though very simple, this parabolic equation has lot of applications in porous media modelling. If it is averaged over the thickness, we obtained the Hele-Shaw relation (Lamb 1906),

$$\overline{v_x} = \frac{1}{h}\int_{-\frac{h}{2}}^{\frac{h}{2}} v_x(z)\,dz = \frac{h^2}{12\mu_\beta}\frac{\Delta P}{L}. \tag{3.13}$$

Hele-Shaw cells consist of two parallel plates fixed at a small distance sandwiching a viscous fluid. They are widely used in fluid mechanics experiments to visualize the flow patterns. In a sense, the microfluidic devices are Hele-Shaw cells with obstacles to mimic a porous medium. This Hele-Shaw equation that relates the average velocity to the pressure gradient reminds us of Darcy's law with a permeability $K = \frac{h^2}{12}$ and a porosity $\phi = 1$. Actually, this simple integration was historically the first step towards the derivation of Darcy's law from Stokes momentum equation (see Vasil'ev (2009) for an interesting history review from Hele-Shaw's first experiments to Darcy's law formulation). We will see in Chapter 5 that the Hele-Shaw equation can be used to solve 2D depth-averaged flow problems in microfluidic devices.

Equation 3.12 also has applications for flow in fractured media. Indeed, at leading order, the fluid flow through fractures is conceptualized by using the assumption of laminar flow between parallel plates. The total flow between parallel plates, $q = \overline{v_x}hw$ is equal to,

$$q = \frac{h^3 w}{12\mu_\beta}\frac{\Delta P}{L}. \tag{3.14}$$

We recognize, here, the cubic law where h represents the fracture aperture.

3.3.2 Viscous flow through a capillary

Another important analytical solution of the Navier-Stokes equations is the Poiseuille flow in a capillary, see Figure 3.7. This solution, first proposed by the physicist and physiologist Poiseuille for the study of blood flows in capillary, has a lot of impact on porous media research since it is at the origin of the well-known Kozeny-Carman correlation to estimate the permeability according to the porosity.

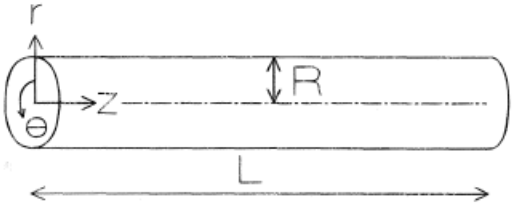


Figure 3.7: Schematic of flow through a capillary

Considering that the tube is long enough compared to its radius and due to axial symmetry, we can assume that $\nabla p = \frac{\Delta P}{L}\boldsymbol{e}_z$ and that the velocity is only carried by the z component that only depends on the radial position, $\boldsymbol{v}_\beta = v_z(r)\boldsymbol{e}_z$. In cylindrical coordinates, the Stokes momentum equation becomes,

$$\mu_\beta \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial v_z}{\partial r}\right) = \frac{\Delta P}{L}, \tag{3.15}$$

with the no-slip boundary conditions at the walls,

$$v_z(r = R) = 0. \tag{3.16}$$

The integration of this boundary value problem leads to the following parabolic profile,

$$v_z(r) = \frac{R^2}{4\mu_\beta}\left(1 - \left(\frac{r}{R}\right)^2\right)\frac{\Delta P}{L}. \tag{3.17}$$

It is the 3D solution of a Poiseuille flow. We notice that the maximum velocity, $v_{z,max} = \frac{R^2}{4\mu_\beta}\frac{\Delta P}{L}$, is reached in the middle of the capillary. The intrinsic average of this equation yields in,

$$\overline{v_z}^\beta = \frac{2}{R^2}\int_0^R v_z\left(r\right) r dr = \frac{R^2}{8\mu_\beta}\frac{\Delta P}{L}. \tag{3.18}$$

The total flow through the capillary tube, $q = \overline{v_z}^\beta \pi R^2$ is equal to,

$$q = \frac{\pi R^4}{8\mu_\beta}\frac{\Delta P}{L}. \tag{3.19}$$

We recognize, here, the Hagen-Poiseuille equation. This equation is the base element of the Pore-Network Modelling (PNM) technique – the historical technique used to simulate flows in porous materials at the pore-scale whose pioneer work is attributed to Fatt (1956) who used the analogy with electrical networks to compute capillary pressure curves. The main idea of the pore-network modelling is to approximate the pore-space by interconnected pore bodies represented by nodes and pore throats represented by bonds in which Hagen-Poisseuille law is applied. Although PNM is appealing because of its simplicity, the technique suffers from two strong limitations. First, the approximation of the pore structure with cylindrical bonds is not always representative of the actual geometry that involves corners and tortuous pathways. Then, the physical laws used in PNM describe averaged processes within the bonds and cannot be used to investigate physical mechanisms that occurs within the bonds. Unlike PNM, l approaches introduced in this thesis (Chap. 5 and Chap. 6) solve directly the Stokes equations in the complex pore structure geometry.

Chapter 4

Scientific strategy: multi-scale analysis of hydro-bio-geochemical processes

The scientific approach adopted throughout my research relies on a modelling strategy involving a cascade of scales nested within each other to develop large-scale models rooted in a correct description of the elementary physical principles including media heterogeneity. Large-scale models are obtained by combining consistent averaging with a fine-scale description of the physicochemical processes using a combination of pore-scale simulations and experiments in well-controlled environments.

4.1 The challenging coupled processes

4.1.1 Multi-physics couplings

Mass fluxes in geological porous media involve processes that relate to very different physics including hydrodynamics, electrostatic, geochemistry, microbiology, and poromechanics. Very often, these physics are strongly coupled and can be treated independently from each other only under assumptions that are most of the time too limiting for describing real systems. A good understanding of the complex feedback between the processes is crucial to quantify the different couplings and assess the dominant mechanisms. The main mechanisms identified in the mass transport in hydro-geosystems are the fluid flow (single and two-phase), transport of particles in the pore space, the chemical reaction at the mineral surface implying dissolution/precipitation and a change in the rock properties, heat transfer, the evolution of electric charges, the deformation of the solid matrix, and the biological activity of microorganisms.

4.1.2 Multi-scale couplings

Modelling processes in heterogeneous geological formations is also a multi-scale problem with characteristic length sizes ranging from the molecular-scale (subnanometer) to the basin scale (km and above). The mathematical description of subsurface processes varies with the modelling resolution, and consequently with the size of the

studied system. Indeed, it is not conceivable – even imagining the most powerful computational resources – to apprehend transport processes in a sedimentary basin with molecular dynamics simulations in which all the interactions between molecules are considered. The classic approach to developing large-scale mechanistic and predictive models rooted in the elementary physical principles is to consider a modelling strategy involving a cascade of scales nested within each other (molecular-scale, pore-scale, core-scale, and field-scale, see Fig. 4.1). The successive scales are coupled between them through boundary conditions and the evolution of constitutive laws. The transition across scales is a crucial step that necessitates sophisticated mathematical tools to interpret the smaller-scale phenomena in terms of upscaled models. Homogenization techniques (e.g. volume averaging (Gray et al. 1993; Whitaker 1999)) and statistical analysis (Dagan 1989; Matheron 1965) are used to bridge the gap between the scales and relate large scale behaviours to microscale physical-chemo processes. Classically, three levels of description are considered: the molecular-scale, the pore-scale, and the so-called Darcy-scale.

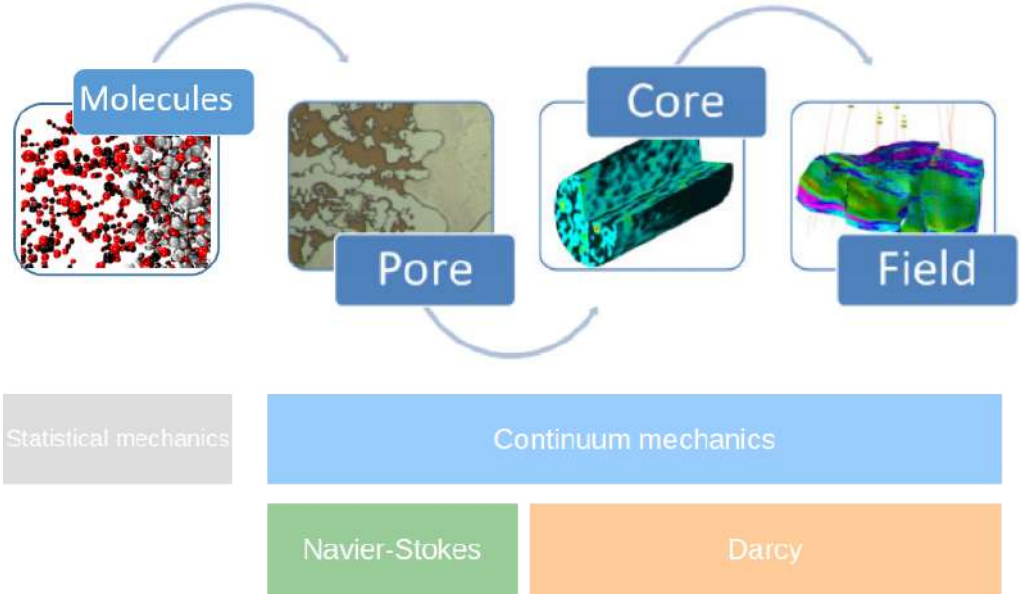


Figure 4.1: Modelling strategy involving a cascade of scales nested within each other (after Buchgraber (2013)).

Molecular scale All the interactions between molecules – fluid and solid – are described explicitly using statistic mechanics. Molecular dynamics relies on the knowledge of bonds between atoms and the force fields applied to them (e.g. electrostatic, van der Waals).

Pore scale Solid and fluid phases are described by continuous media separated by sharp interfaces. Investigations at the pore-scale imply an exact knowledge of the rock pore structure using either microtomography imaging or concept geometries. This is the smallest scale on which continuum mechanics applies. Therefore, Navier-Stokes equations describe the fluid flow in the pore space. The description of the processes at the discontinuity between phases – i.e. between two immiscible fluids

(e.g. air-water, oil-water) or at the fluid-rock interface – is a key aspect of pore-scale modelling and also a major challenge.

Darcy scale The rock microstructure and the mapping of minerals are not described explicitly. Instead, each point of the domain representing the system corresponds to a fluid-solid aggregate. Fluid flow is described by Darcy's law – or a variation – and all the rock topology is lumped into effective parameters including porosity, specific surface area, and permeability. The terminology "Darcy scale" does not refer to a particular physical length but to the smallest scale above which Darcy's law applies. In a logic of a cascade of scales, it is relevant to consider multiple Darcy scales nested within each other. In such a case, for a given scale, the medium heterogeneity is homogenized by introducing effective properties (e.g. porosity, permeability) that depend on the underlying level of description.

4.2 High-resolution studies of hydro-geochemical phenomena

The first step of the multi-scale analysis proposed in this chapter concerns investigations at the pore scale. This requires the development of suitable modelling tools for simulating flow and transport within the pore space. Experiments in well-controlled environments including microfluidics and microtomography allow the acquisition of high-resolution reference data to benchmark and accompany the code development.

4.2.1 *Digital Rock Physics* and computational microfluidics

Improvements in imaging technologies and step changes in computing power are making it possible to image natural porous media with high resolution. Digitized images of the complex pore-space in three dimensions (3D) are often used to analyze the flow dynamics and compute flow-related quantities (see Fig. 4.2), such as permeability, relative permeability, and capillary–pressure relations (Spanne et al. 1994). The process of imaging natural rocks and modelling the flow dynamics using the high-resolution representations of the pore-space is often referred to as *Digital Rock Physics*. This emerging technology is a natural companion of laboratory experiments as it can be used to perform sensitivity analysis for identifying the key parameters of the underlying processes or to explore ranges of conditions including pressure and temperature that are difficult to reach in the laboratory without dedicated equipment. Application areas include reservoir engineering, subsurface hydrology, and subsurface CO_2 sequestration. Specifically, X-ray microtomography (micro-CT) permits 3D imaging of the pore structure of rock samples that are a few cubic millimeters in size at a resolution a micron (e.g., images made up of 2000^3 voxels with a voxel size of one cubic micrometer; Blunt et al. (2013) and Wildenschild and Sheppard (2013)).

Improvement of *Digital Rock Physics* technologies – and their physical engines made of computational microfluidics for geosciences (see Chap. 5) – is mainly driven by the development of appropriate physics and efficient multi-scale algorithms to account for rock heterogeneities and complex coupled processes. The micro-continuum approaches that I develop (Soulaine and Tchelepi 2016) are central in my modelling

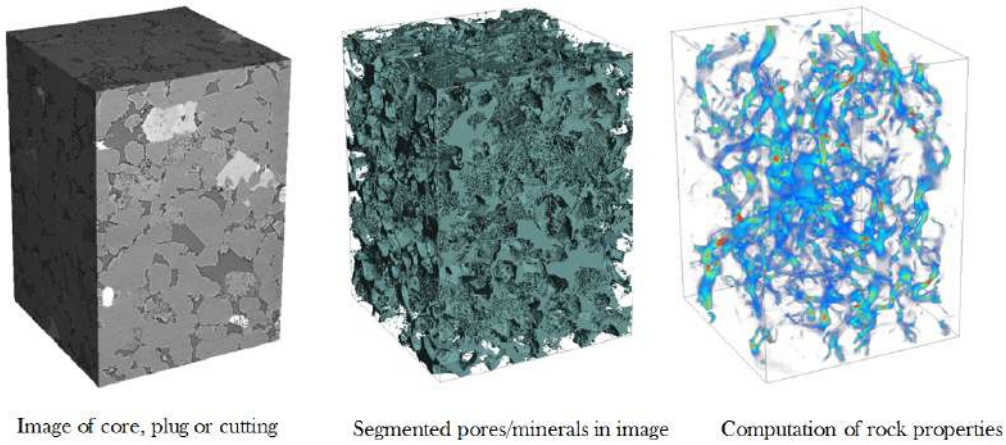


Figure 4.2: Successive steps of the acquisition and manipulation of digital rocks. *a)* Rock sample is imaged with X-ray microtomography. *b)* The image is segmented into pores and solid minerals *c)* Pore-scale physics is solved in the image (here a Stokes flow) and the results are averaged and interpreted in terms of rock properties (e.g. permeability) (source: GeoDict).

strategy. They are two-scale approaches currently able to simulate with great accuracy solute transport, mineral dissolution, multi-phase flow, and matrix deformation (see Chap. 6).

4.2.2 Experiments in well-controlled environments

Experiments in controlled environments allow in-depth investigations of subsurface phenomena by singling a mechanism out of the complex coupled processes. Combined with advanced image processing, they provide quantitive data needed for verifying the computational microfluidics and Digital Rock Physics that I develop.

Microfluidics Microfluidic experiments allow precise control and direct visualization of flows, reactions, and transport mechanisms at the microscale. Microfluidic devices are being used increasingly in the field of geosciences. They are sometimes referred to as Aquifer-on-a-chip or Geological-Lab-on-Chip (GLoC) (Anbari et al. 2018). Historically, they were used to identify two-phase flow regimes in pore networks (Lenormand et al. 1988). Since this pioneering work, the microfabrication techniques improved significantly. Classically, a representation of the pore space is etched on a substrate by photolithography methods and then covered by a transparent material. The etched pattern may be of very variable complexity ranging from single micro-channels of different shapes to patterns representative of a natural porous medium obtained from imaging of real rocks, as illustrated in Figure 4.3. Micromodel surface properties can be controlled (Zhao et al. 2016). Recently, micromodels have been etched directly in carbonate rocks (Song et al. 2014), hence opening new possibilities in studying reactive transport in natural porous media. To achieve control and accurate quantification of the relevant parameters of the processes under study, microfluidic experiments are usually coupled with high-resolution imaging and metrology techniques. In that regard, the nano-microfluidic lab (NanoμLab) at ISTO is equipped with micro-Raman and infrared spectroscopy

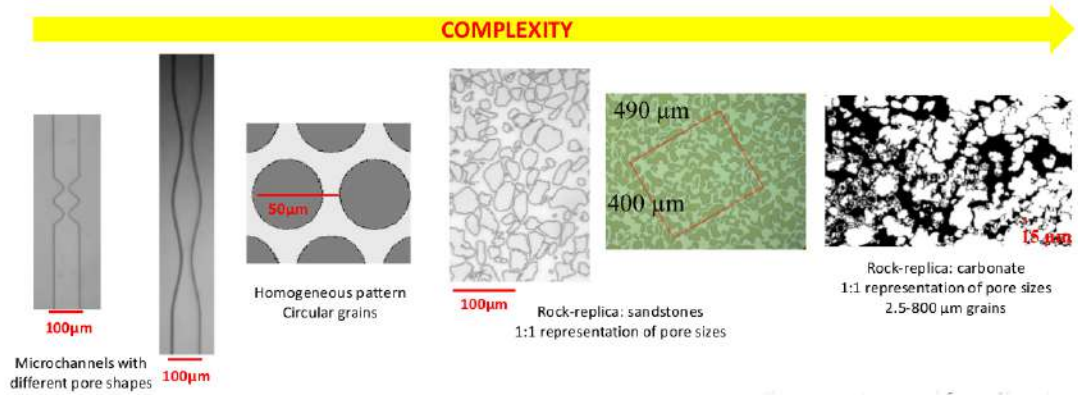


Figure 4.3: Example of micromodel patterns designed and used at the Earth Sciences Institute of Orléans (ISTO), sorted by increasing geometrical complexity (Credit: Dr. Sophie Roman, ISTO).

instruments to monitor the chemical evolution of the system (Poonoosamy et al. 2020) and has strong expertise in velocimetry techniques (Roman et al. 2017, 2016). Moreover, within the University of Orléans, ISTO has a partnership with GREMI (Groupe de Recherches sur l'Energétique des Milieux Ionisés, UMR 7344 Université d'Orléans-CNRS) to fabricate the microfluidics systems in their ISO-4 cleanroom.

High-resolution imaging X-ray microtomography imaging is now an essential tool for observation and characterization of geological microstructure with a resolution up to the microns and below. It is a non-invasive imaging technique that allows for constructing a 3D image of an object using a set of two-dimensional radiographs of the X-ray attenuation properties of the object materials. Some setups offer in situ imaging under experimental conditions in real-time. They can be used to follow the four-dimensional, time-lapse changes of geometry and fluid distribution during dynamic processes or experiments. Applications include multi-phase flow studies (Armstrong et al. 2014; Blunt et al. 2013; Garing et al. 2017), deformation experiments (Renard et al. 2018; Renard et al. 2016), or hydrogeochemical processes (Noiriel and Soulaine 2021). The 3D micro-CT images are also the base element of *Digital Rock Physics* simulations.

4.3 Core and field scales

Upscaling with homogeneisation techniques Mathematical tools such as homogenization techniques are used for changing spatial scale to develop large-scale models used in reservoir simulators. The method of volume averaging is an appealing technique to derive partial differential equations governing the flow and transport phenomena at large scales (Whitaker 1999). The resulting models involve effective parameters including surface area, permeability, dispersion tensors, and transfer functions that describe pore-scale physics and rock geometrical properties. They are evaluated by averaging pore-scale simulations over a representative elementary volume of the rock sample.

Lab-scale simulations and experiments Column-scale experiments (from a few millimeters to decimeters) are used to verify the robustness of upscaled models. Comparisons between simulation and experiment are usually done through breakthrough curves. Probes (pressure, temperature, concentration) along the column length can also be used to establish one-dimensional profiles of the transport processes. For sub-centimeter columns, time-resolved X-ray micro-CT imaging can be used to monitor the temporal evolution of in situ colloid deposition and concentration, fluid distribution and remobilization, and porosity reduction in a non-destructive way (Godinho et al. 2019; Kottsova et al. 2021; Molnar et al. 2014). This way, the concentration and porosity profiles along the column length inform the continuum-scale model development.

Large-scale measurements As the final objective of this multi-scale scientific strategy is to develop predictive capacities at the field scale, the developed numerical models are compared with field-scale data. Here, well-bore analysis (e.g. hydraulic head measurements, push-pull tracer tests, slug tests) provide essential datasets to benchmark the field-scale models. Advanced geophysical techniques provide a 3D mapping of the subsurface processes. These promising approaches are still under development, the main challenge being the deconvolution of the measured signal to identify the signature of hydro-bio-geochemical processes[1].

[1]This is the main objective of the ANR project IMAGE started in 2022 and of the postdoc of Dr. Flore Rembert

Chapter 5

Computational microfluidics for geosciences

Computational microfluidics for geosciences[1] *is the third leg of the scientific strategy that includes microfluidic experiments and high-resolution imaging for deciphering coupled processes in geological porous media. This modelling approach solves the fundamental equations of continuum mechanics in the exact geometry of the porous material. Computational microfluidics intends to complement –and sometimes replace– laboratory experiments. Although the field is still in its infancy, the recent progress in modelling multiphase flow and reactive transport at the pore-scale has shed new light into the coupled mechanisms occurring in geological porous media already. In this chapter, we review the state-of-the-art of computational microfluidics for geosciences, the open challenges, and the future trends.*

5.1 Introduction

Since their appearance in the early eighties, microfluidic experiments have revolutionized the knowledge in porous media research. They brought new insights into the debates that drive the community for decades including the controversial use of Darcy's law for modelling multiphase flows and the understanding of the complex feedback between hydro-bio-chemical systems. The most famous example is the work of Lenormand et al. (1988) who identified different flow instabilities in unsaturated porous systems according to the fluid pair and the flow rates using micromodels. The latter enables direct visualization of the flow, transport, and chemical processes in rock microstructure replica sandwiched between two parallel plates separated by a small distance (Jahanbakhsh et al. 2020; Morais et al. 2020). Coupled with high-resolution imaging both in space and time, microfluidic experiments have shed new light into the key mechanisms controlling subsurface flow. For example, particle image velocimetry has highlighted internal fluid eddies within droplets trapped by capillary forces whose large scale impact is not included in multiphase Darcy's law (Roman et al. 2016, 2020). Microfluidics also led to substantial advancements in the understanding of reactive systems as the most recent microchips involve reactive materials to investigate dissolution and precipitation of solid minerals in fractured porous media (Osselin et al. 2016; Poonoosamy et al. 2020; Song et al. 2014; Soulaine

[1]This chapter in adapted from Soulaine et al. (2021c)

et al. 2017b, 2018). In a few decades, microfluidics has become an indispensable tool in geosciences to decipher the complex mechanisms that occur in porous systems and the discipline keeps improving.

Computational microfluidics is the digital counterpart of microfluidic experiments. As any modelling approach, it is a natural companion of laboratory experiments as it can be used to perform sensitivity analysis for identifying the key parameters of the underlying processes or to explore ranges of conditions including pressure and temperature that are difficult to reach in the laboratory without dedicated equipment. Moreover, computational microfluidics can also assist in the design of micromodels. Unlike pore-network modelling –a widely used approach for describing pore-scale physics (Fatt 1956)– that relies on an approximation of the pore geometries and on pore-averaged physical laws, computational microfluidics solve the fundamental equations of continuum mechanics in the exact microstructure geometry. Historically, this kind of simulation was considered computationally expensive and almost impossible to run on complex and large domains. The enormous progress in high-resolution numerical simulation of complex flows and high-performance computing techniques, however, make it possible to routinely perform computational microfluidics on domains that reach the size of a Representative Elementary Volume. Although it heavily relies on advanced and modern approaches for simulating fluid dynamics, computational microfluidics for geosciences has its own challenges related to the complexity of natural systems including the heterogeneity of geological porous media, the evolving porous microstructure along with geochemical processes, and the complex description of interfacial phenomena at the mineral surfaces (Meakin and Tartakovsky 2009).

The recent improvements in computational microfluidics for geosciences enable the modelling of multiphase flow, reactive transport (Molins et al. 2020), and biofilm growth. A comprehensive review of the state-of-the-art including my own contributions since 2013, as well as the remaining challenges that need to be solved for being truly predictive, are presented in Section 5.3. An essential aspect of my numerical developments concerns the model verification by direct comparison with microfluidic experiments (Roman et al. 2017; Soulaine et al. 2018). In that regard, the fruitful collaboration with Dr Sophie Roman –first at Stanford University and now at ISTO– had demonstrated the robustness of my models. The ability of ISTO's Porous Media Research Group to design well-controlled microfluidic experiments and obtained high-resolution measurements of the velocity fields (Roman et al. 2016), phase distribution, film thickness (Roman et al. 2017), aqueous species concentrations, and mapping in mineral changes (Poonoosamy et al. 2020) is an inestimable aspect of my research for confronting my numerical models with real life.

Exposing simulation results to experimental data is not always an easy task as it raises questions regarding the measurement resolution and also questions on the modelling spatial description. Micromodels are often referred to as "two-dimensional" objects by opposition to real geological objects or their reproduction using 3D printing. This qualification is somewhat imprecise as the micromodels are made of the assembly of two parallel plates and that important physical phenomena occur in the confined space between the plates. This confusion leads many scientists to use two-dimensional models for representing their microfluidic experiments numerically. In most cases, however, 2D simulations are not able to reproduce the experiments. Even in cases for which the micromodel depth is much larger than the smallest

pore-throat diameter the two-dimensional hypothesis breaks down because physical events are not uniform in the thickness. For example, reactive particles can nucleate in the mid-plane or at the solid walls (Beuvier et al. 2015) and instabilities in the liquid films that wet the top and bottom plates can lead to the formation of bubbles in the bulk (Hansen and Toong 1971; Zhao et al. 2018). The systematic usage of 3D models, however, is computationally intensive. To reduce the computational efforts, 2D depth-integrated models are an interesting alternative for solving flows in microfluidic chips. These approaches, however, can have strong limitations. In Section 5.2 we present depth-integrated models for single-phase, two-phase, and species transport, and we discuss their range of validity.

Our definition of computational microfluidics goes beyond the modelling of flow and geochemical processes in micromodels as it can also be applied to complex 3D structures. In this sense, computational microfluidics for geosciences is intimately linked to Digital Rock Physics, an emerging technology concerned with the estimation of effective upscaled parameters (e.g. permeability, dispersion tensor) in three-dimensional images of rock samples (Blunt et al. 2013). In Digital Rock Physics, these properties can be calculated using different techniques, such as pore network analysis (Andrä et al. 2013a,b) or closure problem approaches (Soulaine et al. 2013, 2021a; Whitaker 1999). Alternatively, computational microfluidics can be used to directly solve the flow and transport equations within the pore-space and compute the desired upscaled properties. Although it is often restricted to small micro-CT images ($< 300^3$ voxels) or to single-phase flow due to high computing cost, computational microfluidics is also well suited to investigate pore-to-pore physics (Ferrari and Lunati 2014; Pavuluri et al. 2020) with the objective of improving upscaling techniques (e.g. pore entry pressure for pore network modelling). A major asset of microfluidics is the ability to design well-controlled experiments and obtain high-resolution measurements – both in space and time – of the velocity fields (Roman et al. 2016), phase distribution, film thickness (Roman et al. 2017), aqueous species concentrations (Chang et al. 2017), and mapping in mineral changes (Poonoosamy et al. 2020). Microfluidics is of inestimable value to verify the numerical models using direct comparison with microfluidic experiments (Chapman et al. 2013; Oostrom et al. 2016; Roman et al. 2017; Soulaine et al. 2018; Willingham et al. 2008; Yoon et al. 2012). Therefore, our integrated approach combining computational and experimental microfluidics guarantees the development of robust and validated numerical models for pore-scale modelling and Digital Rock Physics.

This Chapter aims at reviewing the modelling approaches in computational microfluidics for geosciences and their open challenges, as well as my personal contributions to this emerging scientific discipline. It is organized as follows. First, I introduce the fundamental equations used in computational microfluidics (Sec. 5.2.1) and the common numerical approaches to solve them (Sec. 5.2.2). I also present models that solve directly for the depth-averaged variables at a reduced simulation cost (Sec. 5.2.3). Then, I summarise the main advances we have done in modelling multiphase flow (Sec. 5.3.1) and reactive transport (Sec. 5.3.2), and I discuss future lines of research.

5.2 Mathematical models and numerical solutions

In this section, we introduce the fundamental equations used in computational microfluidics for solving flow and transport. We also discuss the standard numerical approaches for solving these equations.

5.2.1 Fundamental governing equations

Computational microfluidics rely on the fundamental equations of continuum mechanics for fluid flow, namely the Navier-Stokes equations, solved in the exact geometry of the pore space. They are composed by a set of mass and momentum balance equations combined with boundary conditions at the solid surface.

Flow equations For single-phase flow, the conservation of mass reads,

$$\frac{\partial \rho}{\partial t} + \nabla . \left(\rho \boldsymbol{v} \right) = 0, \tag{5.1}$$

where ρ and $\boldsymbol{v}$ are the fluid density and velocity, respectively. For liquids, it is common to assume that the fluid is incompressible and the mass balance equation reduces to the divergence-free velocity,

$$\nabla . \boldsymbol{v} = 0. \tag{5.2}$$

In a Eulerian frame, the momentum balance reads,

$$\frac{\partial \rho \boldsymbol{v}}{\partial t} + \nabla . \left(\rho \boldsymbol{v} \boldsymbol{v} \right) = -\nabla p + \rho \boldsymbol{g} + \nabla . \mu \left(\nabla \boldsymbol{v} + \nabla^t \boldsymbol{v} \right), \tag{5.3}$$

where p is the pressure field, $\boldsymbol{g}$ is the gravity, and μ is the fluid viscosity. The left-hand side corresponds to the inertial effects. As mass flow rate in the soils and the subsurface are usually low, it is common to neglect this term and to use Stokes instead of the Navier-Stokes momentum equation. On the right-hand side, the first term is the pressure gradient, the second is the gravity acceleration and the third term is the viscous dissipation. The latter characterizes the internal friction of the fluid particles with each other. The higher the viscosity, the larger the pressure gradient has to be to initiate the fluid movement. At the pore-scale, it is also common to neglect the gravity term.

Alternative forms of the Navier-Stokes equations including non-Newtonian and compressible fluids as well as description of the equations in a Lagrangian frame are found in any fluid mechanics textbook (e.g. Caltagirone (2013) and Landau and Lifshitz (1987)).

Boundary conditions at the solid walls The flow equations are supplemented by boundary conditions at the solid surface that arise from particle/solid molecular interactions. For viscous flow, it is common to consider that the fluid molecules adhere to the surface due to molecular interactions including van der Waals or Coulombic forces. Because of this adherence, the fluid particles at the solid wall

cannot slide on the surface and the tangential component of the velocity is null, leading to the *no-slip* condition,

$$\boldsymbol{v} = 0. \tag{5.4}$$

The no-slip boundary condition is valid for most of the Newtonian liquids flowing in porous media with pore-throat larger than 100 nm. Although widely used in fluid dynamics, the no-slip condition is not always valid in geological systems. In particular, in complex fluids involving colloids or polymers, the fluid elements adjacent to the surface can travel along the surface by rolling or sliding. In such cases, the fluid in contact with the solid surface slide over it with a finite value. Moreover, the no-slip condition poses a problem in viscous flow theory at the place where an interface between two fluids meets a solid boundary. Instead, the so-called *partial slip* condition,

$$\boldsymbol{v} = \beta \left(\mathsf{I} - \boldsymbol{nn}\right) . \left(\boldsymbol{n}. \left(\nabla \boldsymbol{v} +^{t} \nabla \boldsymbol{v}\right)\right), \tag{5.5}$$

proposed by Navier (1823) and later revisited by Maxwell (1879), relates the tangential velocity to the strain rate. In this relation, $\boldsymbol{n}$ is the normal vector at the solid surface and β is called the slip length and is typically of the order of ten nanometers.

Solute transport In its simplest form, the transport of a species i in a fluid phase is modelled by an advection-diffusion equation:

$$\frac{\partial C_i}{\partial t} + \nabla . \left(\boldsymbol{v} C_i\right) = \nabla . \left(D_i \nabla C_i\right), \tag{5.6}$$

where the first term is the accumulation term, the second term is the advection term and the right-hand side corresponds to diffusion effects where D_i is the diffusion coefficient of species i into the fluid phase. More complex forms of the solute transport includes multi-component diffusion, thermal diffusion and electrostatic gradients.

As for flow equations, the solute balance equation is supplemented by boundary conditions at the solid walls. For a passive tracer the *no flux* condition writes,

$$\boldsymbol{n}. D_i \nabla C_i = 0. \tag{5.7}$$

If the species reacts with the solid surface the *reactive flux* condition is,

$$\boldsymbol{n}. \left(-D_i \nabla C_i\right) = r_m \left(C_i\right), \tag{5.8}$$

where r_m is a source or sink term that describes kinetic reactions or adsorption phenomena.

It is also common to consider that the species is in *thermodynamic equilibrium* with the solid wall using the condition

$$C_i = C_i^{eq}, \tag{5.9}$$

where C_i^{eq} is a fixed value imposed by thermodynamic equilibrium.

5.2.2 Numerical engines for solving Navier-Stokes equations

Solving Navier-Stokes equations in complex geometries is not trivial, and besides very few situations, there are no analytical solutions. Computational microfluidics

uses numerical approximations to solve the flow within the pore space. Complexities include the pressure-velocity coupling and the non-linearity due to the inertial term in the momentum equation. In this section, we introduce some standard approaches for solving single-phase Navier-Stokes equations. Specific techniques for multiphase flow, reactive transport and particulate transport are introduced and discussed in Section 5.3.

Computational Fluid Dynamics In Computational Fluid Dynamics (CFD), the Navier-Stokes equations are solved by discretizing the spatial differential operators on an Eulerian grid using techniques (see Fig. 5.1) such as the finite difference method (FDM), the finite volume method (FVM), or the finite element method (FEM) (Ferziger and Peric 2002). In principle, the three families of techniques yield the same solution if the computational grid is fine enough. For fluid flow, however, FVM is usually preferred as it is conservative and deals with unstructured grids by construction. In FVM, the domain is subdivided into a finite number of contiguous control volumes whereby the integral form of the conservation laws is applied. Nowadays, all the commercial or open-source CFD simulators embed very efficient Navier-Stokes solvers that can deal with very large grids and complex geometries using multigrid strategies and High-Performance Computing. Solution algorithms usually rely on predictor-corrector methods to solve for the pressure-velocity coupling. The Pressure-Implicit with Splitting of Operators (PISO) algorithm is one of the most popular for solving transient flows (Issa 1985). Algorithms such as Semi-Implicit Method for Pressure Linked Equations (SIMPLE) solve directly for steady-state flows (Patankar 1980).

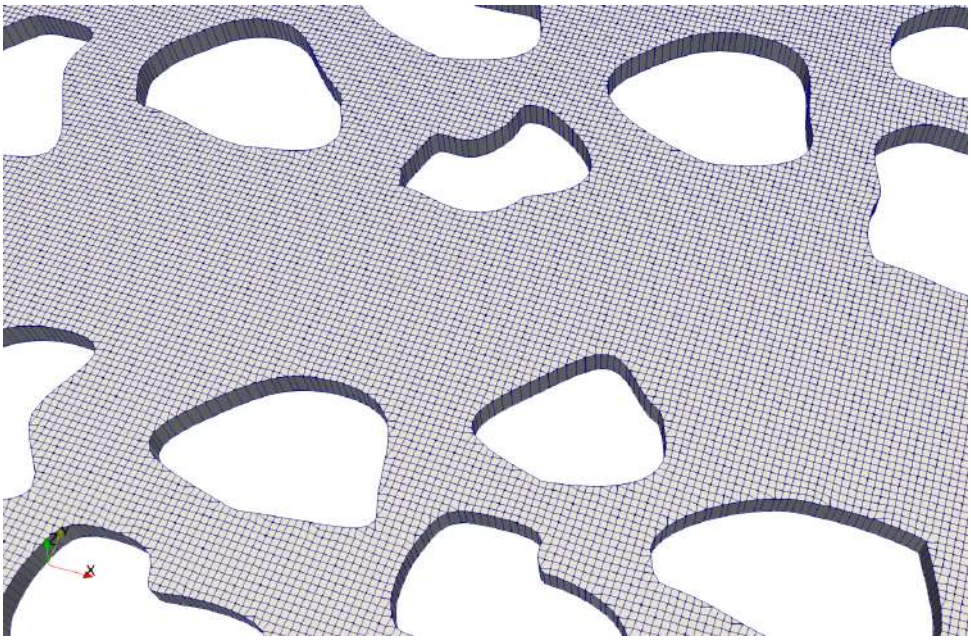


Figure 5.1: Example of finite-volume grid used in computational microfluidics.

Lattice Boltzmann method The lattice Boltzmann method (LBM) is an alternative technique that gains in popularity to solve the flows at the pore-scale. The popularity of LBM comes from the ease to program massively parallel codes.

Unlike traditional CFD approaches that solve for Navier-Stokes equations, LBM is based on the Boltzmann equation that describes the probability of finding a given particle at a given position with a given velocity. This is a statistical approach and the averaged behaviour approximates the continuum mechanics Navier-Stokes equations. In LBM, a discrete Boltzmann equation is solved on a lattice where the particles can only moved along a finite number of directions. The degree of freedom are determined by the nature of the lattice classified with the nomenclature DnQm where n corresponds to the dimension of the lattice (n=2 for two-dimensional simulations, n=3 for three-dimensional simulations) and m stands for number of directions. The Boltzmann equation involves a collision operator that describes the change in the probability distribution function induced by the collisions between particles. In the Lattice Boltzmann method, the boundary conditions are described in terms of probability distribution functions. The description of the wall conditions corresponds actually to the physical model of boundary events proposed by Maxwell (1879). For a no-slip, the bounce-back condition is used where the particles are reflected at the wall. Hence, the net average flow rate at the solid surface is zero. Slip conditions can be obtained using specular reflections (Succi 2002). Partial slip is modeled with a mix of bounceback and specular reflections. The implementation of boundary conditions in LBM can become very complex when the solid surface is not aligned with the lattice.

Smoothed-Particle Hydrodynamics Smoothed-Particle Hydrodynamics (SPH) is a mesh-free modelling technique that describes fluids as a set of discrete moving particles in a Lagrangian framework. The method was initially developed to model the interaction of stars in astrophysics (Gingold and Monaghan 1977; Lucy 1977) and is now being used increasingly in real-time animation and video games. The particles have a spatial distance over which their properties are smoothed by a kernel function centered on the particles. This distance defines the sphere of influence for each particle and is used to reconstruct continuous variables such as fluid density. The SPH particles are then used as interpolation points to discretize and solve the governing equations.

SPH has several benefits over traditional grid-based methods. First, it is meshless and there is no need to go through complex gridding processes. Second, it explicitly conserves mass and momentum. Because the method is Lagrangian, there is no non-linear term in the momentum equation and that allows an accurate description of the advection terms when dealing with high flow rates. Third, the SPH Lagrangian framework allows for the modelling of moving and deformable boundaries without using complex tracking algorithms. SPH has also important drawbacks. Although the SPH algorithms due to their simplicity and structure are easy to program, the implementation of boundary conditions can be tricky. Moreover, for the same accuracy, SPH is more computationally intensive than FVM or FEM because it requires much more neighboring particles than the tensile of grid-based methods. However, because SPH can be easily paralleled, the rise of parallel computing including Graphical Processing Units makes SPH a promising technique (Vacondio et al. 2020).

5.2.3 Depth-averaged models: 2D vs 3D

Most of the time, measurements in microfluidic experiments relate to depth-averaged values. For example, even though micro-Particle Image Velocimetry that uses confocal microscopy or laser pretends to measure the velocity profiles in a cross-section, these techniques do measure values averaged in a thickness of the order of few micrometers that corresponds to the laser thickness or the close surrounding of the focal plane (Roman et al. 2016). Likewise, microfluidic experiments using optical luminescence for measuring a concentration profile (Watson et al. 2019) or mapping the water distribution (Roman et al. 2017; Zhao et al. 2016) provides values that are averaged along the micromodel depth, h, without information of the depth profile of the species concentration or water saturation. The depth-averaged variables are defined as the vertical integration of the 3D fields,

$$\overline{\boldsymbol{v}}^{2D} = \frac{1}{h}\int_0^h \boldsymbol{v}\,(x, y, z)\,dz. \tag{5.10}$$

In this section, we present depth-averaged models that directly provide the solution for the depth-averaged velocity, $\overline{\boldsymbol{v}}^{2D}$, pressure, $\overline{p}^{2D}$, and concentration, $\overline{C_i}^{2D}$, fields without solving 3D problems. These models result from the integration of the fundamental equations described in Section 5.2.1 along the depth axis and can significantly reduce the simulation costs. They have, however, ranges of validity that are discussed below. The following discussion is based on the work we published in Soulaine et al. (2021b).

Single-phase flow: The integration of Navier-Stokes momentum equation over the micromodel depth along with the assumption that a parabolic profile approximates the flow in the thickness gives,

$$\frac{\partial \rho \overline{\boldsymbol{v}}^{2D}}{\partial t} + \nabla.\left(\rho \overline{\boldsymbol{v}}^{2D}\overline{\boldsymbol{v}}^{2D}\right) = -\nabla \overline{p}^{2D} + \nabla.\mu\left(\nabla \overline{\boldsymbol{v}}^{2D} + \nabla^t \overline{\boldsymbol{v}}^{2D}\right) - \mu\frac{12}{h^2}\overline{\boldsymbol{v}}^{2D}, \tag{5.11}$$

where the last term of the right-hand side, $\mu\frac{12}{h^2}\overline{\boldsymbol{v}}^{2D}$, is the Hele-Shaw correction (Lamb 1906), i.e. a drag force related to the friction of the fluid along the micromodel top and bottom plates. Numerical implementation of Eq. (5.11) has been reported using CFD (Roman et al. 2016) and LBM (Laleian et al. 2015; Venturoli and Boek 2006). The Hele-Shaw correction is similar to a Darcy law for which the permeability is $k = \frac{h^2}{12}$ and the porosity is $\phi = 1$. This simple depth-integration was historically the first step towards the derivation of Darcy's law from Stokes equation (Vasil'ev 2009). Actually, for micromodels with a low aspect ratio between the cell thickness and the pore-throat size, $h/d \ll 1$, the drag force is the dominant force and Eq. 5.11 can be replaced by Darcy's law for Hele-Shaw cells,

$$\overline{\boldsymbol{v}}^{2D} = -\frac{h^2}{12\mu}\nabla \overline{p}^{2D}. \tag{5.12}$$

Theoretically, the hypothesis that flow sandwiched in between two parallel plates obey a parabolic profile is only valid for infinite plates – their dimensions are much larger than the space between them. Micromodels mimicking rock porous structures, however, contain pillars that violate this assumption with three-dimensional

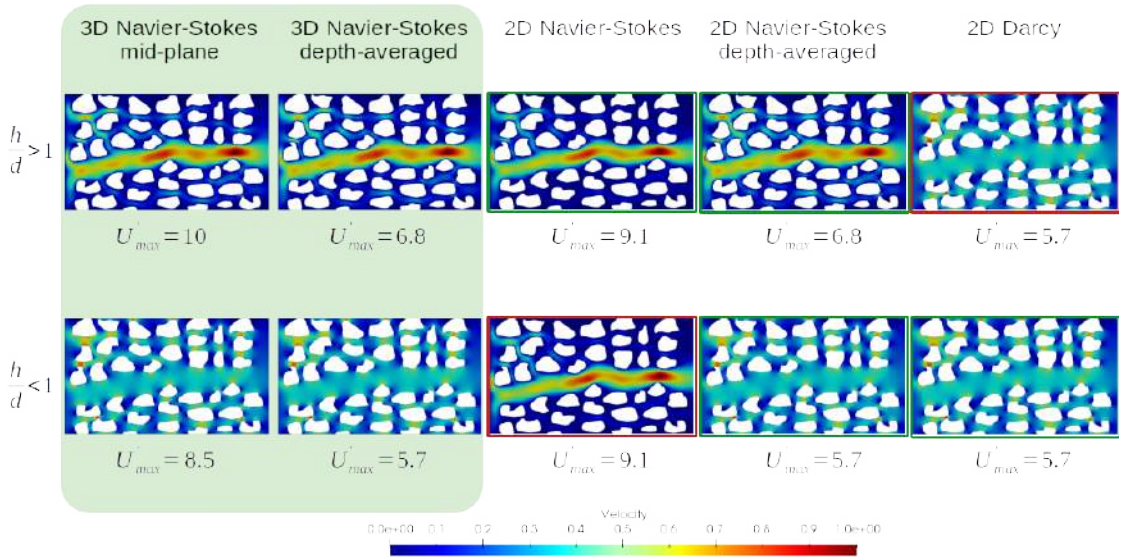


Figure 5.2: Single-phase flow simulations using 3D, 2D and 2D-depth-averaged models for different aspect ratio between the micromodel thickness, h and the smallest pore-throat, d. The computational domain is gridded with 1.4×10^5 cells for the 2D cases and $11.4 \times 10^5 \times 10$ for the 3D cases. The typical pore-throat size is $d_{typ} = 4 \times 10^{-3}$ m, the largest is $d_{large} = 12 \times 10^{-3}$ m and the smallest is $d = 0.5 \times 10^{-3}$ m. We used two different depth thickness, namely $h = 10^{-2}$ m and $h = 10^{-4}$ m. Simulations are performed with OpenFOAM. For each simulation, the velocity profile is normalized by the maximum value, and we indicate the maximum value normalized by the inlet velocity. If $\frac{h}{d} > 1$, 2D Navier-Stokes simulations tend toward the mid-plane 3D Navier-Stokes solution and 2D Darcy is far from the reference solution. If $\frac{h}{d} < 1$, 2D Darcy captures the 3D reference solution whereas 2D Navier-Stokes breaks down. The 2D depth-integrated Navier-Stokes model captures the 3D flow behaviour regardless the micromodel thickness (from Soulaine et al. (2021b)).

flowlines at the vicinity of the corner formed by the pillars and the top and bottom plates. Nonetheless, in practice, Eq. (5.11) is a very good approximation of the flow behaviour in Hele-Shaw cells. In Roman et al. (2016), the very good agreement in the comparison of the velocity profiles obtained experimentally with micro-PIV and numerically solving Eq. (5.11) illustrates that the depth-averaged flow model is relevant for micromodels. This statement is verified numerically in the simulation results presented in Figure 5.2 for which the velocity profile in a micromodel was computed based on 2D, 2D depth-integrated, or 3D simulations for different micromodel thickness. For 3D simulations, the 2/3 factor in the velocity magnitude of the 3D depth-integrated profile with respect to the mid-plane value is representative of a parabolic profile in the third dimension. The 2D Navier-Stokes model only tends to the 3D solution if $h/d > 1$ and corresponds to the 3D mid-plane profile. In this case, the scales separation hypothesis leading to Darcy's law is not satisfied and the 2D Darcy solution cannot describe the flow through the micromodel. If $h/d < 1$, the friction of the fluid along the top and bottom plates are the dominant resistance to the flow. In such a case, the 3D flow tends to the 2D Darcy solution whereas the 2D Navier-Stokes solution – that corresponds to an infinite thickness – is completely

off the reference data. The 2D depth-averaged simulations remain consistent with the 3D solution regardless of the aspect ratio.

Single-phase scalar transport: The integration of an advection-diffusion equation for the transport of a scalar –e.g. species concentration or temperature– across the micromodel thickness gives,

$$\frac{\partial \overline{C}^{2D}}{\partial t} + \nabla.\left(\overline{\boldsymbol{v}}^{2D}\overline{C}^{2D}\right) = \nabla.\left(\mathsf{D}(t).\nabla\overline{C}^{2D}\right), \tag{5.13}$$

where $\overline{\boldsymbol{v}}^{2D}$ is obtained using Eq. (5.11), and $\mathsf{D}(t)$ is a time-dependent dispersion tensor. Because of the parabolic velocity profile in the micromodel depth, there are hydrodynamic dispersion effects that must be considered even if the aspect ratio between the pore-throat size and micromodel thickness is lower than one, $h/d > 1$. For transport in between two parallel plates, the dispersion tensor exactly writes (Dentz and Carrera 2007),

$$\mathsf{D}(t) = D_i\mathsf{I} + \frac{h^2}{4D_i}\overline{\boldsymbol{v}}^{2D}\overline{\boldsymbol{v}}^{2D}\left(\frac{2}{105} - \sum_{n=1}^{\infty}\frac{18}{(n\pi)^6}\exp\left(-4\left(\frac{n\pi}{h}\right)^2 D_i t\right)\right), \tag{5.14}$$

where the first term is the molecular diffusion and the second term corresponds to Taylor-Aris dispersion in the direction of the flow (Aris 1956; Taylor 1953) – note that $\overline{\boldsymbol{v}}^{2D}\overline{\boldsymbol{v}}^{2D}$ is a tensor. The time-dependent infinite series accounts for the pre-asymptotic regime at early time and tends towards zero at long time. In practice, only the two first terms of the series are necessary.

Hydrodynamic dispersion effects are characterised by the Péclet number, $Pe_h = \frac{hU}{D}$, that compares the transport by advection and the transport by diffusion. They are illustrated in the simulation results of Fig. 5.3 that solve the transport of a scalar using 3D, 2D, and 2D depth-averaged models for Péclet numbers ranging from 0.1 to 10^4. The deviations between mid-plane and depth-averaged values of the 3D simulations illustrate the hydrodynamic dispersion when advection is the dominant transport mechanism. Experimentally, the measurement of species concentration using optical index corresponds to depth-averaged values (Watson et al. 2019). Confusion regarding the nature of measured value may lead to misinterpretations of the comparison between experiments and 3D simulations. For transport dominated by diffusion, $Pe_h < 1$, the concentration profile is flat over the micromodel depth. Therefore, there are no dispersion effects and the 2D models are in good agreement with 3D simulations. If the advection takes over, $Pe_h > 1$, hydrodynamic dispersion becomes important and 2D model breaks down. The two-dimensional depth-averaged simulations using the time-dependant dispersion tensor approximate well the depth-integrated solution of the 3D simulations, regardless the Péclet number. Figure 5.4 highlights the importance of considering the pre-asymptotic regime. Neglecting the dispersion (i.e. $\mathsf{D}(t) = D_i\mathsf{I}$) or considering only the asymptotic Taylor-Aris dispersion (i.e. $\mathsf{D}(t) = D_i\mathsf{I} + \frac{h^2\overline{\boldsymbol{v}}^{2D}\overline{\boldsymbol{v}}^{2D}}{210D_i}$) tends to overdisperse the solute. Likewise, it is crucial to consider the product $\overline{\boldsymbol{v}}^{2D}\overline{\boldsymbol{v}}^{2D}$ as a tensor and not as a scalar $\left|\overline{\boldsymbol{v}}^{2D}\right|^2$ because Taylor-Aris dispersion in microfluidic devices is not isotropic and occurs in the direction of the flow.



Figure 5.3: Transport of a scalar in a micromodel with aspect ratio $h/d > 1$. We use 3D, 2D and 2D-depth-averaged models for different Péclet number by varying the diffusion. The results correspond to isocontour of the concentration for $C = 0.2$ (blue), $C = 0.5$ (green), $C = 0.8$ (orange) at $t = 42\,\text{s}$. 2D models wihout dispersion break down for $Pe_h > 10$ (from Soulaine et al. (2021b)).

Two-phase flow Simulation of two-phase flow processes in microfluidic cells using 2D depth-averaged models is challenging as the fluid-fluid interface has two curvature radii: one in the micromodel plane, another in the micromodel thickness. Horgue et al. (2013) proposes 2D depth-averaged equations in which they integrated the interface curvature along the micromodel thickness in addition to the Hele-Shaw correction. Their model, however, is limited to cases for which no liquid films are flowing on the top and bottom plates, and cannot capture phenomena such as snap-off. Alternatively, Cueto-Felgueroso and Juanes (2014) proposes a Darcy-like model to simulate two-phase flow in Hele-Shaw cells that accounts for water films flowing on the top and bottom plates using relative permeabilities. The applicability of their model, however, is restricted to aspect ratios between the micromodel thickness and the pore-throat size smaller than one, $h/d \ll 1$. In absence of reliable 2D depth-integrated models for two-phase flow, it is recommended to use 3D rather than 2D simulations since the former can capture the impact of wetting films along the top and bottom plates as well as corner flows (Zhao et al. 2019). Moreover, phenomena such as snap-off can only be captured if the two curvature radii are considered

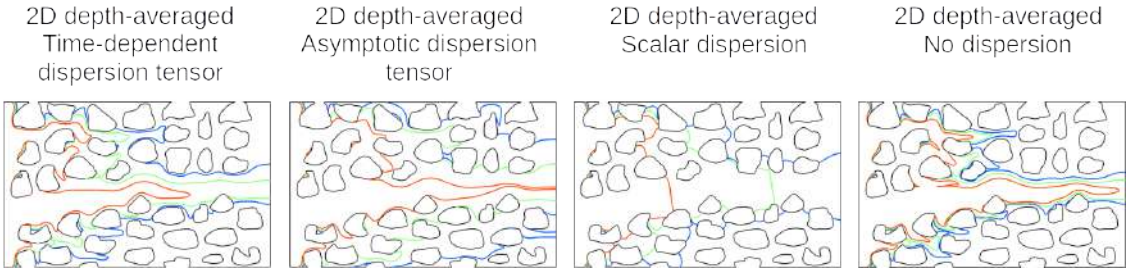


Figure 5.4: Comparison of the transport of a scalar at $Pe = 10^3$ in a micromodel with aspect ratio $h/d > 1$ using 2D depth-averaged models including time-dependent dispersion tensor, the asymptotic dispersion tensor, isotropic dispersion and no dispersion. The results correspond to isocontour of the concentration for $C = 0.2$ (blue), $C = 0.5$ (green), $C = 0.8$ (orange) at $t = 42\,\mathrm{s}$. Neglecting the dispersion (i.e. $\mathsf{D}(t) = D_i\mathsf{I}$) or considering only the asymptotic Taylor-Aris dispersion (i.e. $\mathsf{D}(t) = D_i\mathsf{I} + \frac{h^2\overline{\boldsymbol{v}}^{2D}\overline{\boldsymbol{v}}^{2D}}{210D_i}$) tends to overdisperse the solute. Isotropic dispersion cannot capture the transport of scalars in micromodels (from Soulaine et al. (2021b)).

(Roman et al. 2017). It is among my future work to derive an depth-integrated model for two-phase flow. The micro-continuum model for two-phase flow that we published with Francisco Carrillo and Ian Bourg (Carrillo et al. 2020) has the potential to combine the models of Cueto-Felgueroso and Juanes (2014) and Horgue et al. (2013) and therefore to come up with a more efficient depth-integrated model for two-phase flow.

Reactive transport There is no universal depth-averaged equations for solving reactive systems as three-dimensional effects are important. For example, nucleation effects occur either at the wall surfaces or in the bulk depending on a wide variety of parameters including pH, salinity and surface roughness.

5.2.4 Multi-scale approach

Bi-modal pore-size distributions are quite common in geological porous media. This special feature appears, for example, in porous fractured media for which the width of a fracture is orders of magnitude larger than the typical pore throat diameter in the porous matrix. In such a case, a full Navier-Stokes modelling approach requires to resolve all the porosity explicitly and leads to tremendous computational grid size. Alternative approaches propose to model only flow in the fracture using Navier-Stokes while the flow in the matrix is described by Darcy's law.

In a cumulative and continuous effort started in 2013, I am developing the concept of micro-continuum modelling (Carrillo et al. 2020; Soulaine et al. 2019, 2016, 2017b, 2018; Soulaine and Tchelepi 2016), i.e. an intermediate approach between a pure Navier-Stokes description of the transport for which all the porosity is fully resolved and a pure continuum-scale modelling based on Darcy's law (Soulaine and Tchelepi 2016). It relies on a hybrid-scale flow model based on the Darcy-Brinkman-Stokes equation (Brinkman 1947; Neale and Nader 1974) discretized with the Finite-Volume Method. All the geometric structures below a threshold that corresponds to the spatial resolution of the simulation is filtered and modelled as a porous medium.

The momentum equation reads

$$\frac{1}{\phi}\left(\frac{\partial \rho \overline{\boldsymbol{v}}}{\partial t} + \nabla.\left(\frac{\rho}{\phi}\overline{\boldsymbol{v}}\overline{\boldsymbol{v}}\right)\right) = -\nabla\overline{p} + \nabla.\frac{\mu}{\phi}\left(\nabla\overline{\boldsymbol{v}} + \nabla^t\overline{\boldsymbol{v}}\right) - \mu k^{-1}\overline{\boldsymbol{v}}, \tag{5.15}$$

where the overline notation denotes filtered variables, ϕ is porosity field, and k is the permeability of the porous regions. Our recent studies show that the micro-continuum framework is well suited to describe the movement of fluid/solid interfaces caused by dissolution/erosion or precipitation/deposition (Molins et al. 2020; Soulaine et al. 2017b; Soulaine and Tchelepi 2016) and to solve problems involving two characteristic length scales (Carrillo et al. 2020; Soulaine et al. 2016, 2021c). The next chapter is devoted to a comprehensive review of the micro-continuum models.

5.3 Application to geosciences and open challenges

In this section, I present my contributions in the field of computational microfluidics for geosciences since 2013. I review the main advances we have made in modelling multiphase flow and reactive transport at the pore-scale. I also discuss the open-challenges I identified and my current and future lines of research in this area.

5.3.1 Modelling of immiscible two-phase flow

Modelling immiscible multi-phase flow is one the biggest challenge in the porous media community. Despite a very large usage in subsurface engineering, many studies have emphasized the limits of Darcy's law for two or more fluids (Cinar et al. 2009; Danis and Quintard 1984; Rose 2000). An accurate description of the flow mechanisms in porous media is challenged by the complex interplay between capillary, viscous, and gravitational forces that may lead to highly unstable flows and trapping mechanisms. The heterogeneous nature of geological formations further complicates the development of predictive models.

Computational microfluidics for multiphase flows offers an appealing framework to probe the underlying mechanisms in unsaturated porous media. Despite enormous improvements, however, the numerical modelling of multi-phase flow remains challenging. The difficulties are twofold: on the one hand, numerical methods must be improved (for calculating the interface curvature dynamically, for tracking the composition of the fluids, for speeding-up the simulation times), and on the other hand the physical description of interfacial properties is still not well-established (e.g. wettability conditions, thin films). My research intends to address these open-challenges by developing reliable and efficient multi-phase flow simulators at the pore-scale.

Efficient numerical solvers for capillary-dominated flow The dynamic modelling of multiphase flow phenomena at the pore-scale is complex because it involves the tracking of multiple interfaces that evolve as a function of hydrodynamic forces including gravitational effects, viscous forces, and surface tension forces. Fast events including interface break-up or coalescence and pore invasion by abrupt jumps

(known as Haines jumps) require robust numerical solvers. Furthermore, the modelling is complicated by the dynamics of the contact line or thin films along the solid walls.

I started to look at efficient two-phase flow solvers during my postdoc and the PhD of Moataz Abu AlSaud at Stanford University in the group of Prof Hamdi Tchelepi. The standard approaches for tracking the interface movement of two immiscible fluids rely on solving single-field equations for the single-field variables. Whether the modelling is grid-based or particle-based, a phase indicator function maps the distribution of the fluids within the pore-space and evolves as the fluid-fluid interface propagates along with hydrodynamic forces. This function denotes either the fluid volume fraction in every grid cells for the Volume-of-Fluid (VOF, Hirt and Nichols (1981)) and phase-field approaches, a distance to the fluid interface for Level-Set (LS, Sussman et al. (1994)) approaches, or a color function for Smooth Particles Hydrodynamics (SPH, Tartakovsky and Meakin (2005)) and Lattice Boltzmann Methods (LBM, Gunstensen et al. (1991)). The phase indicator function is used to define single-field variables by weighting the fluid properties (e.g. viscosity, density) according to the distribution of fluid phases. Its gradient is also used to calculate the interface curvature. Today, none of the existing approaches can solve two-phase flow problems accurately and efficiently. One of the bottlenecks is the existence of spurious velocities near the interface attributed to inaccuracies in the calculation of the interface curvature, regardless of the computational approach. This problem acknowledged for more than 30 years (Brackbill et al. 1992), is particularly limiting for capillary-dominated flows (Scardovelli and Zaleski 1999) that are the dominant regimes in many subsurface systems (Cinar and Riaz 2014). An important part of my research in computational microfluidics for two-phase flow consists in developing robust solvers that minimize these numerical issues.

The models I am developing are based on an algebraic VOF approach, i.e. the phase indicator, α_l, is described by the volume fraction of fluid within each cell of the computational domain as illustrated in Figure 5.5 and the governing equations are obtained by volume averaging the Navier-Stokes equations over a control volume (Maes and Soulaine 2020). For incompressible fluids, the continuity equation writes,

$$\nabla . \boldsymbol{v} = 0, \tag{5.16}$$

the mass balance equation for the wetting fluid is

$$\frac{\partial \alpha_l}{\partial t} + \nabla . \left(\alpha_l \boldsymbol{v} \right) + \nabla . \left(\alpha_l \alpha_g \boldsymbol{v}_r \right) = 0, \tag{5.17}$$

and the momentum equation reads

$$\frac{\partial \rho \boldsymbol{v}}{\partial t} + \nabla . \left(\rho \boldsymbol{v} \boldsymbol{v} \right) = -\nabla p + \rho \boldsymbol{g} + \nabla . \left(\mu \left(\nabla \boldsymbol{v} + {}^t \nabla \boldsymbol{v} \right) \right) + \boldsymbol{F}_c, \tag{5.18}$$

where $\boldsymbol{v}_r$ and $\boldsymbol{F}_c$ are sub-grid models that describe the fluid-fluid interactions in cells that contain the interface. They represent the relative velocity and the surface tension force due to the curvature of the interface, respectively. Although other methods such as level-set (Abu AlSaud et al. 2017, 2018; Sussman et al. 1994) can provide a more accurate description of the sharp interface, the VOF method is attractive due to its flexibility, robustness in terms of mass conservation, and adaptability to more complex physics (e.g. multi-component mass transfer). Because the

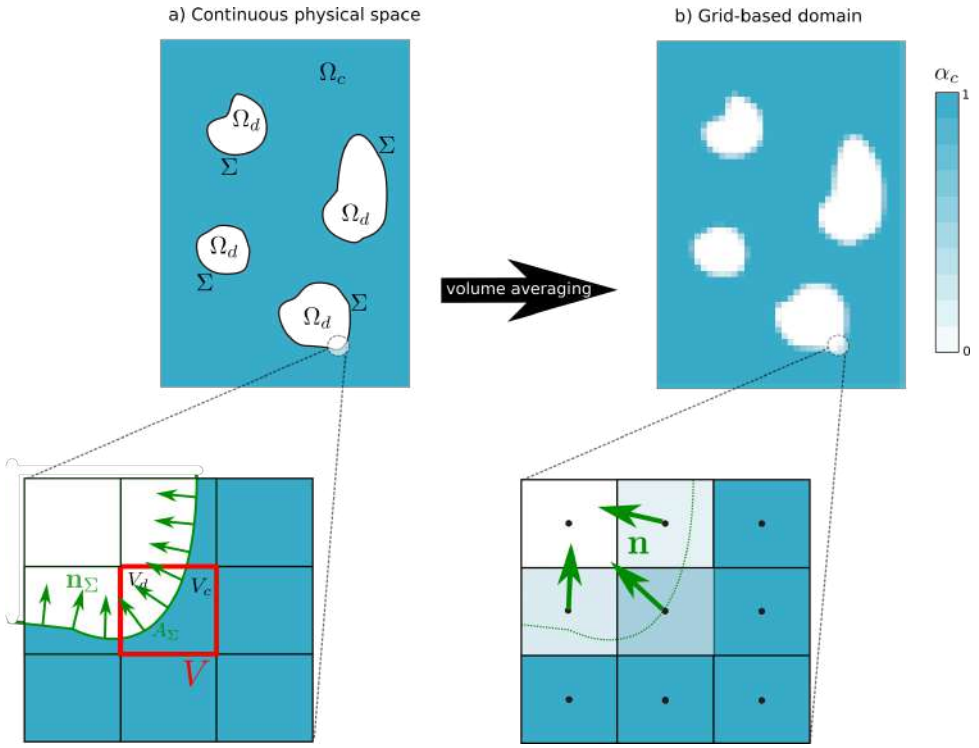


Figure 5.5: Illustration of the volume of fluid approach. It consists of a grid-based approach in which all the properties are volume averaged on a grid block. The volume fraction of fluid in the computational grid is used to track the fluid-fluid interface displacement in the pore-space. Because the exact location of the interface within a cell is not known, VOF uses sub-grid models to describe the fluid-fluid interactions (from Maes and Soulaine (2020)).

derivation of the governing equations relies on volume averaging principles, algebraic VOF methods are the cornerstone of our multi-scale solvers for two-phase flow (Carrillo et al. 2020; Soulaine et al. 2019, 2018).

The two lines of research that I am currently exploring are *(i)* the improvement of the numerics, i.e. the reduction of the numerical errors and a better time-stepping strategy, and *(ii)* a better description of the wettability conditions including the effect of pH and salinity.

Modelling of wettability and interfacial properties Wettability plays a key role in multiphase flow in porous media (De Gennes 1985). Accurate prediction of multiphase flow in the soils and the subsurface for geo-environmental issues including Carbon Capture and geological Storage, water resources remediation, or Enhanced Oil Recovery requires an in-depth understanding at the pore-scale of the key mechanisms that influence the fluid displacements and dynamics including viscous and capillary forces but also the solid surface wettability. Recent studies have shown that the spatial distribution of wettability play a crucial role (AlRatrout et al. 2018; Yesufu-Rufai et al. 2020). The rock wettability and the resulting multiphase flow can be modified by a change in pore water composition (pH, salinity) and interfacial physicochemical properties. For example, an oil droplet trapped within a pore may be more easily remobilized using low salinity water (Jackson et al. 2016). Accurate modelling of wettability conditions at the mineral surface is therefore crucial to

develop efficient and reliable computational microfluidics for two-phase flow.

Traditionally, wettability is described considering that the fluid-fluid interface and the solid surface form a contact angle (see Fig. 5.6). The value of the contact angle is provided from static measurements that described the contact angle at the nanoscale. In computational microfluidics, the cell size near the solid surface is usually orders of magnitude larger than nanometers. Therefore, upscaled laws including the Cox-Voinov model estimates an apparent dynamic contact angle that depends on the interface velocity near the solid surface and accounts for viscous bending (Cox 1986; Voinov 1976). The alteration of interfacial and solid surface properties due to a change of pH and salinity requires constitutive laws for modifying the contact angle accordingly (Maes and Geiger 2017). Moreover, flow models based on contact angles break down in the presence of thin water films on the mineral surface (Starov et al. 2007).

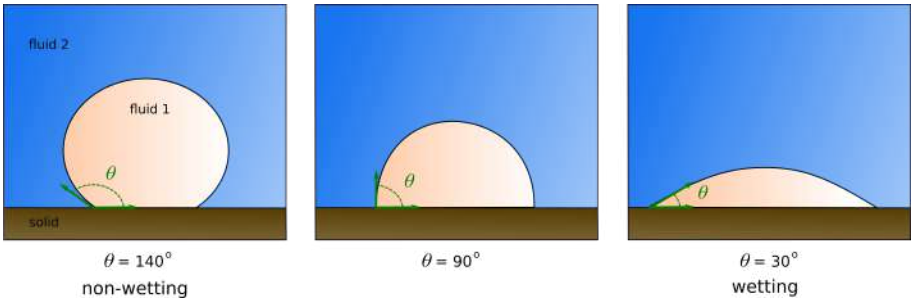


Figure 5.6: The contact angle, θ, is the angle formed by the tangent to the solid surface and the tangent to the fluid-fluid interface. For values smaller than 90°, a droplet lying on a solid substrate tends to spread over the surface. The fluid within the droplet is the wetting fluid with respect to the other fluid and the solid surface. For values larger than 90°, the droplet is the non-wetting phase and tends to minimize its surface area in contact with the solid substrate.

Alternative flow models including lubrication theory offer an appealing framework to account for physicochemical mechanisms controlling the wettability of rocks according to pore water chemistry and surface roughness for instance (Abu AlSaud et al. 2020, 2017). Instead of using a contact angle, lubrication theory provides a partial differential equation governing the evolution of the film thickness, δ, on the solid surface including molecular forces (Pahlavan et al. 2018). It reads,

$$\frac{\partial \delta}{\partial t} + \frac{1}{\mu_w} \nabla . \left[\left(\frac{\delta^3}{3} \nabla \left(\sigma \kappa - \Pi \left(\delta \right) \right) \right) + \frac{\delta^2 \tau}{2} \right] = 0, \tag{5.19}$$

where μ_w is the water viscosity, σ is the surface tension, κ is the interface curvature, $\Pi\left(\delta\right)$ is the disjoining pressure term, and τ is the shear-stress exerted on the film surface. The disjoining –or Derjaguin– pressure includes attractive (e.g. van der Waals) and repulsive (e.g. electrostatic) forces that change with the ionic strength, i.e. pH and salinity. The film equation is solved on the solid surface and is coupled with the two-phase Navier-Stokes solver in the volume of the pore. By getting rid of the concept of contact angle, it is believed that simulators using lubrication models are more realistic and mechanistic for the description of multiphase systems in geological formations for energy and environmental issues. Applications include the modeling of wettability alteration measured in supercritical CO_2 stored in deep

saline aquifers (Wan et al. 2014) and the detachment of crude oil from mineral surface observed when changing the brine from high to low salinity (Berg et al. 2010). The state-of-the-art computational microfluidics using lubrication theory, however, is limited to flat solid boundaries (Abu AlSaud et al. 2020, 2017). Recent work considers curvilinear surfaces on cylindrical solids (Qin et al. 2020). However, pore-scale multiphase flow solvers with lubrication models on generic solid geometries are still under development.

The objective of the PhD thesis of Mojtaba Norouzisadeh who joined our research team in October 2020 is to investigate the physico-chemical mechanisms controlling the wettability of rocks according to pore water chemistry and interfacial properties. We aim at modelling the alteration of interfacial properties due to a change of pH and ionic strength. We are investigating the implementation of lubrication models in the VOF framework to develop more realistic and mechanistic pore-scale multiphase transport solvers in geological formations for energy and environmental issues. This framework will pave the way to the pore-scale modelling of multiphase flow including molecules adsorption and colloids attachment on interfaces. This longer term objective will improve the understanding of colloidal control of immiscible two-phase flow in geological formations and will help to design efficient remediation of contaminated groundwater.

Modelling of species transfer across fluid-fluid interfaces We developed a numerical method to simulate interphase mass transfer along with the evolution of the fluid-fluid interface at the pore-scale. In our approach, the thermodynamic equilibrium at the interface is described by a partitioning relationship such as Henry's law (Henry 1803) and the continuity of mass fluxes is guaranteed as the interface evolves in the pore-space due to fluid displacements including drainage and imbibition (see Fig. 5.7). The development of our model spans over a period of 5 years (2015–2020) with the strong involvement of Marguerite Graveleau (then a Master student at Stanford University) and Dr Julien Maes (a post-doctoral fellow at Heriot-Watt University).

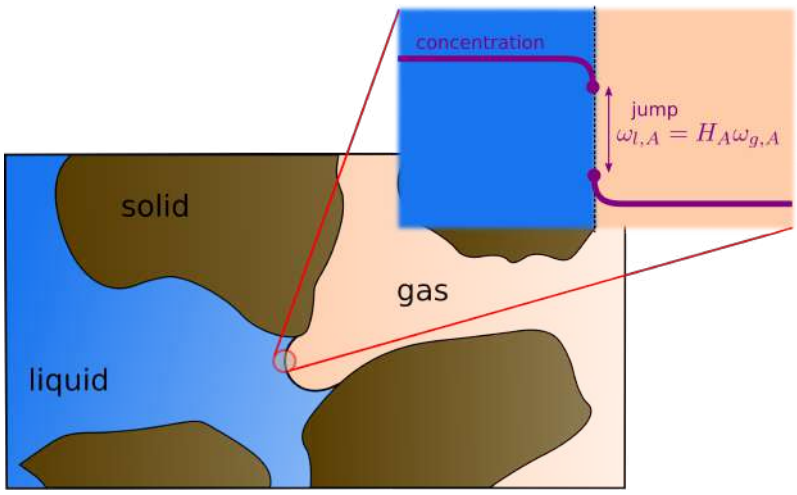


Figure 5.7: Illustration of multi-component mass transfer in two-phase flow at the pore-scale. Thermodynamic equilibrium at the fluid/fluid interface obeys a partitioning relationship such as Henry's law

I started to look at efficient numerical techniques to track the fluid composition in two-phase pore-scale systems when I was a post-doctoral research associate in Prof. Hamdi Tchelepi's group in the Department of Energy Resources Engineering at Stanford University during the Master of Marguerite Graveleau (Graveleau 2016). We were interested in modelling pore-scale mechanisms associated with the injection and sequestration of supercritical carbon dioxide (sCO_2) into deep geological formations. We implemented the Continuous Species Transfer (CST) approach developed by Haroun et al. (2010) into the open-source Computational Fluid Dynamics platform OpenFOAM. CST is a single-field approach along the same lines as the Volume-Of-Fluid technique used for simulating immiscible two-phase flow, i.e. a unique partial differential equation solves for a single-field concentration field regardless the content of a grid block. It reads,

$$\frac{\partial C_A}{\partial t} + \nabla.\left(\boldsymbol{v} C_A\right) = \nabla.\left(D_A^* \left(\nabla C_A + \Phi_A\right)\right), \tag{5.20}$$

where D_A^* is the weighted average of the diffusion coefficients in both fluids and Φ_A is the CST flux enforcing the concentration discontinuity due to Henry's law. It turned out that Haroun's model could not handle situations for which the contact angle between the fluids and the solid surface was different than 90°. In Graveleau et al. (2017), we derived a boundary condition for the CST and used our approach to upscale the rate of mass transfer in unsaturated porous media as a function of the flow rates. Yang et al. (2017), however, pointed out that when convection dominates diffusion locally near the interface, the CST method generates large numerical errors. They show that in order to capture the species concentration discontinuity accurately, the mesh should be extremely refined at the vicinity of the interface. This workaround, however, increases dramatically the computational cost, especially for advection-dominated systems. With Dr Julien Maes, a post-doctoral research fellow at Heriot-Watt University, we carefully revisited the derivation of the CST using the method of volume averaging (Maes and Soulaine 2018). We identify a missing term in CST that was causing unbalanced mass transfer and we came up with a new approach –Compressive-CST or simply C-CST– that is able to simulate interphase mass transfer without limitation on the transport regimes or the grid refinement level.

Our approach can simulate the mass transfer of a passive scalar in a dynamic system (see Figure 5.8). By averaging pore-scale results, we evaluated the mean rate of mass transfer in unsaturated porous media and we proposed new laws that highlight the dependency of the rate of mass transfer in terms of saturation, flow rates and interfacial area. In particular, we are interested in evaluating the role played by the internal re-circulation within the trapped droplets that we observed experimentally (Roman et al. 2016, 2020) on the mass transfer across immiscible interfaces. Moreover, the ability to track the composition of fluids in two-phase flow is crucial for modelling efficiently the change of wettability conditions with pH evolution (Maes and Geiger 2017).

In more recent work, we extended the C-CST for modelling cases for which the species transfer across the fluid-fluid interface is no longer passive but leads to local volume changes (Maes and Soulaine 2020). Such situations happen, for example, when a soda can is opened, the pressure in the headspace falls abruptly and Henry's law is no longer satisfied. To reestablish equilibrium, the carbon dioxide concentration in water decreases by forming gas bubbles. During their travel toward

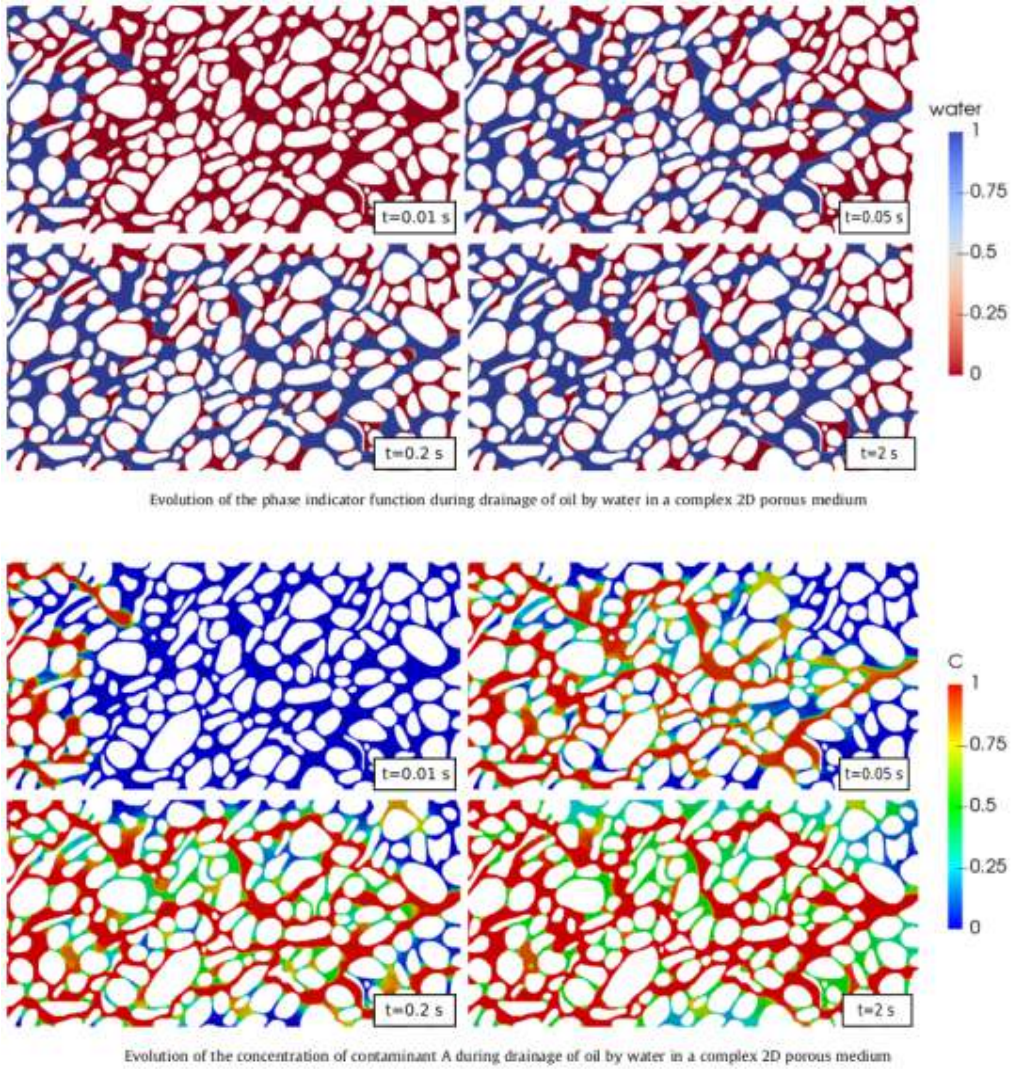


Figure 5.8: Simulation results of drainage of oil by water with multi-component mass transfer at the pore-scale. The white pillars correspond to the solid grains. *Top:* distribution of the fluid phases during the drainage at different timestep. *Bottom:* concentration profile in the system (adapted from Maes and Soulaine (2018)).

the surface, gas bubbles grow because of the diffusive flux from the carbon dioxide dissolved in the liquid (Power et al. 2009). The other way around occurs in carbon sequestration processes when supercritical CO_2 is stored in deep saline aquifers by capillary effects and trapped sCO_2 droplets dissolve into the surrounding brine (Kim et al. 2012). As a consequence, the volume of droplets decreases, and the equilibrium of hydrodynamic forces that was maintaining the sCO_2 trapped in the pore-space is displaced, eventually leading to a mobilization of the droplets. Our C-CST model for phase change can be used to estimate the volume of CO_2 trapped in the porous structure and to assess the timescale of potential remobilization. We are currently in the process of verifying the robustness of our numerical predictions using microfluidics experimental data (see Fig 5.9). Indeed, the micro-nanofluidic platform at ISTO managed by Dr Sophie Roman and Dr Aneta Slodczyk is equipped with a micro-Raman spectrometer that allows local measurements of the concentration evolution within the fluids.

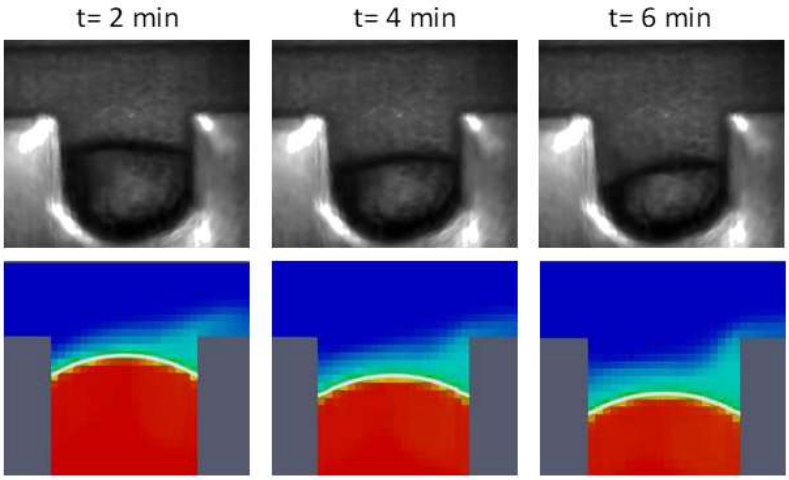


Figure 5.9: Preliminary results of the comparison between experimental and computational microfluidics for the dissolution of trapped CO_2 droplet into water. ISTO micro-nanofluidic laboratory is equipped with a micro-Raman spectrometer that allows point measurements of the fluid composition. This experimental dataset will be used to verify the robustness of our C-CST model for phase change (courtesy of Dr Julien Maes and Dr Sophie Roman).

Nevertheless, the assessment of the fate of an isolated droplet is not enough to fully characterize CO_2 storage by capillary trapping. In a bulk, two neighboring bubbles will interact with each other through a diffusive flux going from the smaller bubble (that has a higher pressure) to the larger bubble. As a consequence, the larger bubbles grow at the expense of the smaller ones. This phenomena relates to the coupling of solubility and interface curvature, a phenomena referred to as Ostwald ripening and well-known in many industrial processes and in the formation of crystals (Lifshitz and Slyozov 1961; Ostwald 1897). Many scientists, however, think that this phenomena is negligible in unsaturated porous media because the bubble shape is constrained mostly by the pore geometry and that the solubility dependence on interface curvature is less important (Li and Yortsos 1995). In future work, we will combine C-CST along with the fluid equations of state to simulate Ostwald ripening. Hence, we will be able to shed light on the long-term debate concerning the existence of Ostwald ripening in porous media and improve the risk assessment related to CO_2 storage in deep saline aquifers.

Multi-scale two-phase flow We have successfully derived, implemented, tested and verified a multiscale model for two-phase flow in porous media. The modelling framework can be used to simultaneously model multiphase flow at two different length scales: *(i)* a Darcy-scale where sub-voxel fluid-fluid and fluid-solid interactions within a porous medium are modelled through relative permeability and capillary constitutive models, *(ii)* a pore-scale (or Navier-Stokes scale) where the solid material is non-porous and fluid-fluid interactions are described through a continuum representation of the Young-Laplace equation.

This unique framework consists in an extension of the micro-continuum approach for modelling two-phase flow in two-scale porous media. Micro-continuum approaches are discussed in-depth in the next Chapter. As of now, it is the only

solver able to simulate two-phase flow in dual systems at the pore-scale. The model development was a stepwise process. The first model I developed combines the Volume-of-Fluid approach for tracking the fluid-fluid interface in regions free of solid and the Darcy-Brinkman-Stokes equation for modelling flow in porous regions (Soulaine et al. 2019). We were able to simulate the expulsion of methane stored in microporous matrix into a soaked fracture (see Figure 5.10). Our results highlight two completely different expulsion flow regimes whether the fracture boundaries are hydrophobic or hydrophilic. On the one hand, we observe a continuous gas production, and on the other hand we obtain a bubbly regime because the surface tension effects at the wet fracture boundary generate an energetic barrier that tends to form gas bubbles as far as the pressure in the microporous regions is sufficient to inflate them. This kind of simulations can be used to assess the rate of hydrocarbon recovery in shale gas according to the spatial distribution of the kerogen (hydrophobic) and clay (hydrophilic) in the vicinity of the micro-cracks (Soulaine et al. 2019).

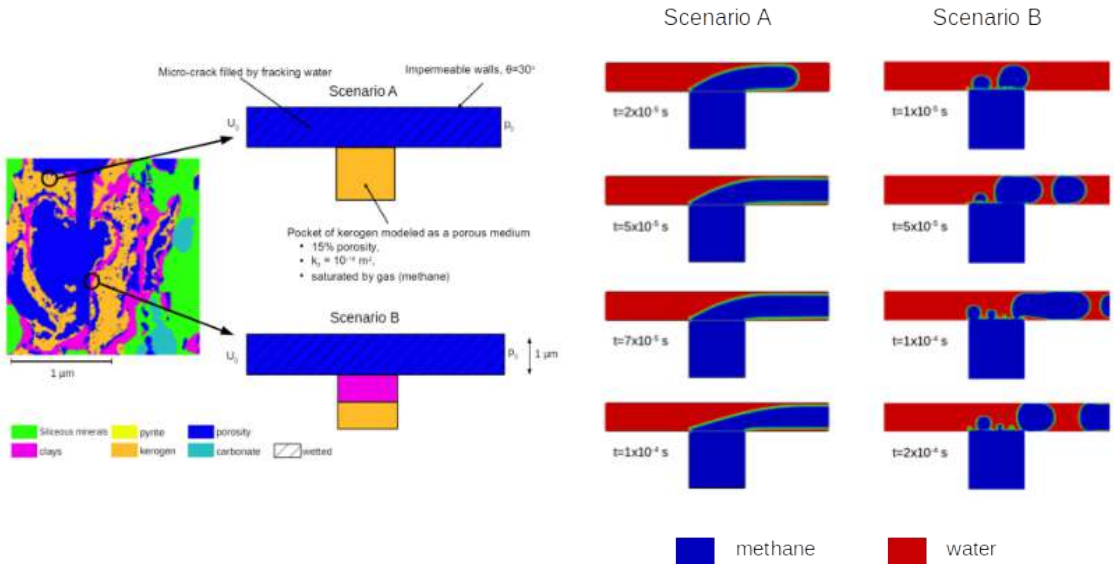


Figure 5.10: Multi-scale simulations of the expulsion of methane stored in a microporous matrix into a soaked fracture. We investigated two scenarios: *A)* the fracture/matrix interface is non-wetting and the methane production is continuous, and *B)* the fracture/matrix interface is wetting and the gas is produced in a bubbly flow regime (adapted from Soulaine et al. (2019)).

Actually, the micro-continuum approach for two-phase flow proposed in Soulaine et al. (2019) oversimplifies the flow physics in the porous domains. In particular, the model could not describe the interplay of gravity and capillarity within the microporous matrix. We lifted this limitation during the PhD thesis of Francisco Carrillo at Princeton University in which we revisited thoroughly the derivation of the model using the method of the volume averaging and asymptotic matching (Carrillo et al. 2020). The model includes classic concepts such as saturation, relative permeability, and capillary pressure curves in the porous matrix. Our multiscale approach offers new possibilities in computational microfluidics to investigate multiphase flow in fractured porous media including the fracture-matrix interactions and to investigate the role played by microporosity in processes such as imbibition and drainage.

Although the proposed formulation represents a significant advance in the sim-

ulation of multiscale multiphase systems, we note that further study is required in particular to properly and rigorously model the multi-scale porous interface. The implemented interface, as it stands, has been shown to accurately predict single phase flow into porous media (Neale and Nader 1974) and impose static contact angles over porous boundaries (Horgue et al. 2014). However, its accuracy when modelling multiphase flow at rough porous interfaces is still an open question, as there does not currently exist a rigorous formulation to model such behaviour. We will use new microfluidic devices involving two characteristic length-scales to probe the interfacial conditions and propose new laws.

Three-phase flow Computational microfluidics for three-phase flow (e.g. gas-water-oil) are still scarce in the literature because the community focuses mostly on two-phase flow simulators for which the open challenges discussed in previous sections are still limiting the predictive capabilities. In the literature, Bayestehparvin et al. (2015) use a Volume-of-Fluid approach, Helland et al. (2017) and Mohammadmoradi and Kantzas (2017) use quasi-static displacement using a Level-Set method, Tartakovsky and Panchenko (2016) use Smooth Particle Hydrodynamics, Dinariev and Evseev (2016) use a phase-field method and Jiang and Tsuji (2017) and van Kats and Egberts (1999) use a lattice-Boltzmann method to investigate three-phase Navier-Stokes flow at pore scale. The development of a robust and efficient three-phase flow simulator at the pore-scale is part of my long-term objectives. Such a tool will be crucial to understand fully the underlying mechanisms of non-aqueous phase liquid (e.g. hydrocarbon, chlorinated solvents) trapped in the unsaturated zone by capillary effects and to propose innovative remediation strategies.

5.3.2 Reactive Transport Modelling at the pore-scale

Most of computational microfluidics developments so far have been devoted to solve the Navier-Stokes equations under single (Bijeljic et al. 2013; Guibert et al. 2015; Soulaine et al. 2016; Spanne et al. 1994) and two-phase flow conditions (Graveleau et al. 2017; Horgue et al. 2013; Maes and Soulaine 2018; Pavuluri et al. 2020; Raeini et al. 2014). Flows in geological porous media, however, have the distinctive feature to interact chemically with the solid walls. The consideration of surface reactions in computational microfluidics requires to include comprehensive reaction networks along with flow and transport, an approach known as Reactive Transport Modelling (Steefel et al. 2005). Despite the growing investment in the development of RTM at the pore-scale – pioneer simulators date back to the late 90s (Békri et al. 1997, 1995) – the field is still in its infancy. The main challenge consists in moving the fluid/solid boundary with respect to chemical reactions at the mineral surfaces (see Fig. 5.11). A large variety of methodologies has been proposed by the community to solve this problem and a comprehensive review can be found in Molins et al. (2020). They include Level-Set (Molins et al. 2017, 2014), or phase-field (Xu and Meakin 2008, 2011) techniques in a Eulerian grid, LBM (Chen et al. 2014c; Kang et al. 2003; Prasianakis et al. 2013; Szymczak and Ladd 2009), SPH (Tartakovsky et al. 2007) and Arbitrary Lagrangian-Eulerian (Oltéan et al. 2013; Starchenko et al. 2016) frameworks. The method I am developing for displacing the mineral interface relies on another family of approaches referred to as micro-continuum (Soulaine et al. 2021c, 2017b; Soulaine and Tchelepi 2016).

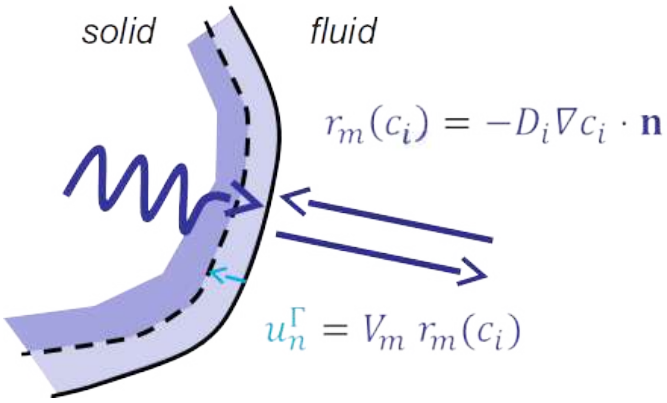


Figure 5.11: Illustration of a moving fluid/solid interface condition: a diffusive flux reacts with the solid mineral and based on the mass balance the interface recedes (dissolution) or advances (precipitation) (Courtesy of Dr Sergi Molins).

My interest in pore-scale modelling of reactive transport dates back to my post-doctoral position at Stanford University and my involvement in the Center for Nanoscale Controls on Geologic CO_2. The objective was to improve the fundamental understanding of the pore-scale processes associated with the injection and sequestration of CO_2 into the subsurface and to assess the long-term issues such as the efficiency of CO_2 retention in reservoir rocks resulting from capillary and dissolution trapping, and from the conversion of dissolved CO_2 to solid carbonate. I developed computational microfluidics to simulate calcite dissolution under single and two-phase flow conditions (Soulaine et al. 2017b, 2018). The simulator was used to investigate the highly non-linear permeability-porosity relationships and to assess the reactive surface area according to flow rates and mineral reactivity. At ISTO, I continue working on the code development with the assistance of Dr Saideep Pavuluri, a postdoc who joined the team in November 2019. All the numerical developments are conducted in concert with microfluidic experiments by Sophie Roman to verify the reliability of the models.

Modelling of dissolution processes I developed a numerical model for simulating dissolution processes at the pore-scale (Soulaine et al. 2017b). In a first step, the surface reaction was described by a first order kinetic. Although this kind of surface reaction is not representative of the complex reaction networks that occur in geological porous media, it allows us to focus on the numerical implementation of fluid/solid moving boundaries. The model we developed relies on an asymptotic behaviour of the micro-continuum technique – a technique discussed in-depth in the next Chapter. Basically, a color function maps the distribution of the solid mineral in an Eulerian grid and moves according to surface reactions. In cells that are free of solid, the flow is modelled by Navier-Stokes equations, and in cells full of solid, the velocity is dropped to near zero value. The surface reaction depends on the fluid-solid interfacial area in a grid cell. The latter is computed using the gradient of the color function. This technique can be qualified of a Volume-of-Solid approach by analogy with the Volume-of-Fluid approach used for simulating two-phase flows and discussed in Section 5.3.1.

We verified the reliability of our computational approach by comparing the simulation results with the high-resolution data of a well-controlled microfluidic exper-

iments that we designed for the occasion (Soulaine et al. 2017b). The setup consists in a calcite crystal posted in a microchannel in which an aqueous solution containing 0.05% of hydrochloric acid is injected at constant flow rate. The evolution of the crystal shape is recorded and analyzed as shown in the top row of Fig. 5.12. The elongated profile of the crystal as it dissolved in the acid flowing solution illustrates well the complex coupling between streamlines, solute transport and surface reaction. It denotes a non-uniform distribution of the solute along the solid surface, leading to faster dissolution rates upstream than downstream. Our model was able to reproduce the shape evolution of the dissolving crystal with great fidelity and without any adjusting parameters. Later, we used the microfluidic data set to benchmark a wide variety of techniques for simulating pore-scale dissolution (Molins et al. 2020). This benchmark exercise gathered five international teams using different techniques including micro-continuum, Level-Set, LBM, moving grids with conformal mapping, and Vortex methods. It turned out that all the simulators were able to reproduce the evolution of the crystal shape, leading to the strong conclusion that pore-scale technologies for moving fluid-solid boundaries along with geochemical reactions are now mature enough to be used as predictive tools.

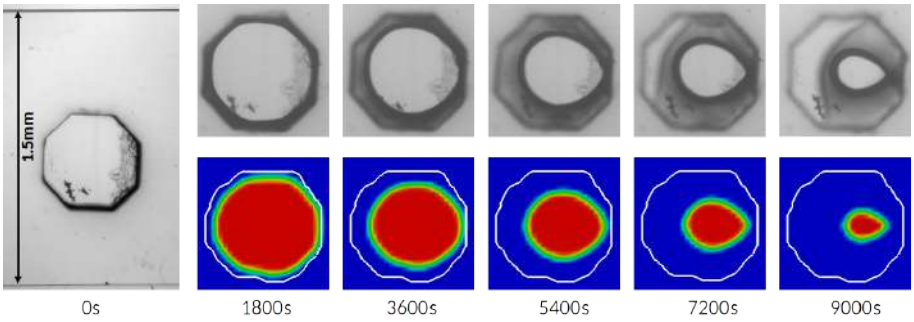


Figure 5.12: Experimental (top) and computational (bottom) microfluidics images of calcite dissolution process at different time steps during the injection of 0.05% HCl at a mean velocity of 1.16×10^{-3} m/s flow rate from the left to the right (from Soulaine et al. (2017b)).

We used our model to investigate the dissolution of micro-model pore networks. Five different dissolution regimes are observed according to the constant of reaction at the mineral surface and the injection mass flow rate as illustrated in Fig. 5.13. For a given fluid, these two effects are characterized by the Damköhler number Da_I and the Péclet number Pe, respectively. These regimes are: compact dissolution ($Pe < 10^{-2}$ and $Da_I > 1$), conical dissolution ($1 > Pe > 10^{-2}$ and $Da_I > 1$), dominant wormhole ($10 > Pe > 1$ and $Da_I > 1$), ramified wormholes ($Pe > 10$ and $Da_I > 1$) and uniform dissolution ($Da_I < 1$ and regardless the Péclet number). These dissolution regimes are confirmed by other studies (Molins et al. 2017; Szymczak and Ladd 2009). Macroscopic properties can be obtained by averaging the pore-scale simulation results. Hence, we showed that in dissolving systems the permeability variations are nonlinear with the injection mass flow rate and that our approach can be used to proposed new permeability-porosity relationships. Moreover, we propose a new law for quantifying the accessible reactive surface area, A, based on the transport and reactivity conditions. It reads

$$\frac{A}{A_0} = 1 - \exp\left(-Pe^{-n} Da_I^{-m}\right), \tag{5.21}$$

where A_0 is the geometric surface area of the porous sample and n and m are model parameters. Although this relation has been obtained heuristically, it fits very well with our pore-scale numerical data. Interestingly, other groups have reconciled their experimental or numerical data used such a formula (Deng et al. 2018; Wen and Li 2017). The analytical solution for the shape evolution of the calcite crystal derived by Prof Piotr Szymczak and his team using conformal mappings could be a good starting point to demonstrate Eq. 5.21.

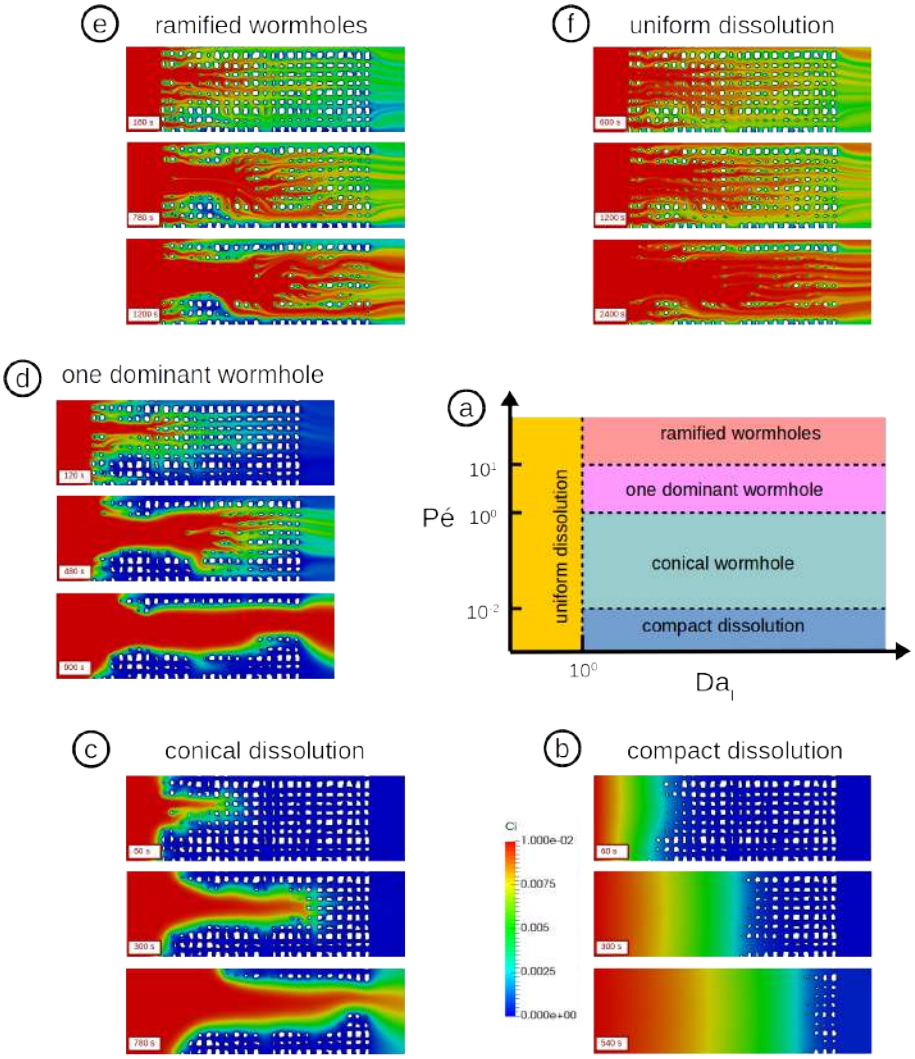


Figure 5.13: Prediction of the evolution of the pore space of a micromodel for different values of the constant of reaction and of the diffusion coefficient (from Soulaine et al. (2017b)).

This first effort was based on a relatively simple geochemical reaction (a single component that reacts with a single mineral using a first-order kinetics) to demonstrate the ability of current codes to simulate mineral dissolution. Further developments and verification still need to be done for simulating multi-component aqueous solutions interacting with heterogeneous multi-mineral media using comprehensive reaction networks. These are part of the research we are conducting with the post-doctoral position of Saideep Pavuluri.

Multi-scale Naturally occurring porous media involve a wide range of spatial scales. For example, the important contrast in pore-size distributions in fractured porous rocks comes from characteristic lengths for the fractures much larger than for the surrounding porous matrix. Therefore, the domain size required to reach a REV limits the use of pure pore-scale modelling. Hybrid-scale models have been proposed to describe systems that include multiple characteristic length-scales for which some regions are described using pore-scale modelling while others are modelled with continuum approaches (Liu and Ortoleva 1996; Liu et al. 1997). Two kinds of approaches solve hybrid-scale problems. On the one hand, the domain decomposition technique solves different physics on separate domains – one for Darcy flow, another for Stokes flow – linked together through appropriate boundary conditions (Molins et al. 2019). On the other hand, micro-continuum models use a single set of partial differential equations throughout the computational domain regardless of the content of a grid block (Soulaine and Tchelepi 2016; Steefel et al. 2015).

Multi-scale models can be also used to investigate the transfer of mass and the development of a weathered zone at the interface between a fracture and a porous matrix in a reactive environment. These are the lines of research we are exploring in the postdoc of Saideep Pavuluri. Theses models are indeed powerful tools to complement and interpret microtomography images of the dissolution or sealing of a fracture (Ajo-Franklin et al. 2019; Noiriel et al. 2007). In that regards, the recent work we published in collaboration with the Institute of Energy and Climate Research (IEK), Jülich Forschungzentrum (Poonoosamy et al. 2020) bring an interesting dataset to investigate hydro-geochemical processes including dissolution and precitipitation in fractured porous media.

Two-phase flow in reactive environment The dynamical modelling of multiphase flow phenomena in reactive environments is quite complex because it involves tracking multiple interfaces that evolve as a function of the details of the reactive transport: on the one hand the fluid/fluid interface is subject to surface tension forces, on the other hand the fluid/solid interface moves with chemical reactions at the surface of the solid minerals. The modelling is complicated by the dynamics of the contact line or thin films along the solid walls, and by the transfer of mass across interfaces. Few research works have been devoted to the modelling of multiphase flow with solid boundaries that evolve with chemical reactions. Parmigiani et al. (2011) have proposed an LBM approach to study the evolution of capillary fingers in a porous medium that dynamically evolves with melting of the solid phase. Later, LBM has been extended to multicomponent multiphase fluid flow and applied to reactive transport with dissolution and precipitation (Chen et al. 2013, 2015). LBM-based approaches, however, are unstable for high density ratios between the liquid and the gas (Chen et al. 2014b), which, therefore, limits the predictive aspects of such modelling frameworks. Importantly, none of these frameworks was verified using experiments.

In Soulaine et al. (2018), we proposed a new pore-scale model to simulate mineral dissolution in unsaturated porous media. The model combines the micro-continuum approach developed in Soulaine et al. (2017b) along with a Volume-of-Fluid technique to track the fluid/fluid interface including surface tension forces. Chemical reactions and wettability conditions at the mineral surface are described as immersed boundaries that evolve with the dissolution of the solid phase. Along the

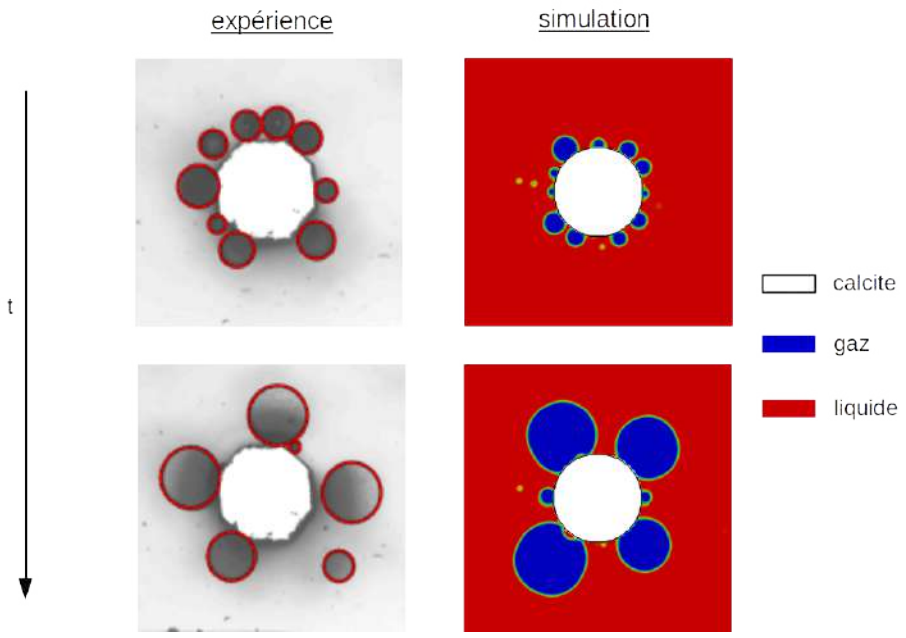


Figure 5.14: Comparison between experimental and computational microfluidics. A calcite crystal is dissolved in a flow of hydrochloric acid at atmospheric conditions. Carbon dioxide gas bubbles nucleate at the mineral surface, grow, coalesce and eventually detach. The micro-continuum model for two-phase flow in reactive environments can reproduce accurately the microfluidic experiments (from Soulaine et al. (2018)).

same lines as our former work regarding the modelling of calcite dissolution at the pore-scale, we used microfluidic experiments to verify the predictive aspect of our model. The experiments consisted in a calcite crystal dissolving in a microchannel after the injection of an aqueous solution containing up to 1% of hydrochloric acid. As the experiments were conducted at atmospheric pressure, a multitude of small gas bubbles nucleate at the solid surface because of the surface reaction. As the bubbles grow, they coalesce with each other and their number decreases significantly until three or four large bubbles are visible. As they keep growing, the gas phase covers a larger surface of the solid and prevents the acid from reacting with the mineral. Eventually, large bubbles detach, and the whole sequence is repeated. Our numerical model was able to capture accurately the sequence of events. In particular, the immersed boundaries used for the wettability conditions and the chemical reactions at the mineral surfaces capture the dynamics of the mineral dissolution under multiphase flow conditions (See Fig. 5.14).

The model we developed is a significant breakthrough in computational microfluidics as it is the first verified pore-scale simulator for reactive multiphase flow in a dissolving porous medium. It has been used to investigate the effect of carbon dioxide production during the acidizing of carbonate formations. Our results show that under single-phase flow conditions, i.e. when the dissolution time of CO_2 into the aqueous phase is instantaneous, the solid structure dissolves forming wormholes. However, if the characteristic time scale of CO_2 dissolving in brine is much longer compared with the rate of gas production, then the simulation results show that CO_2 bubbles grow, coalesce and form flow barriers that limit the transport of the

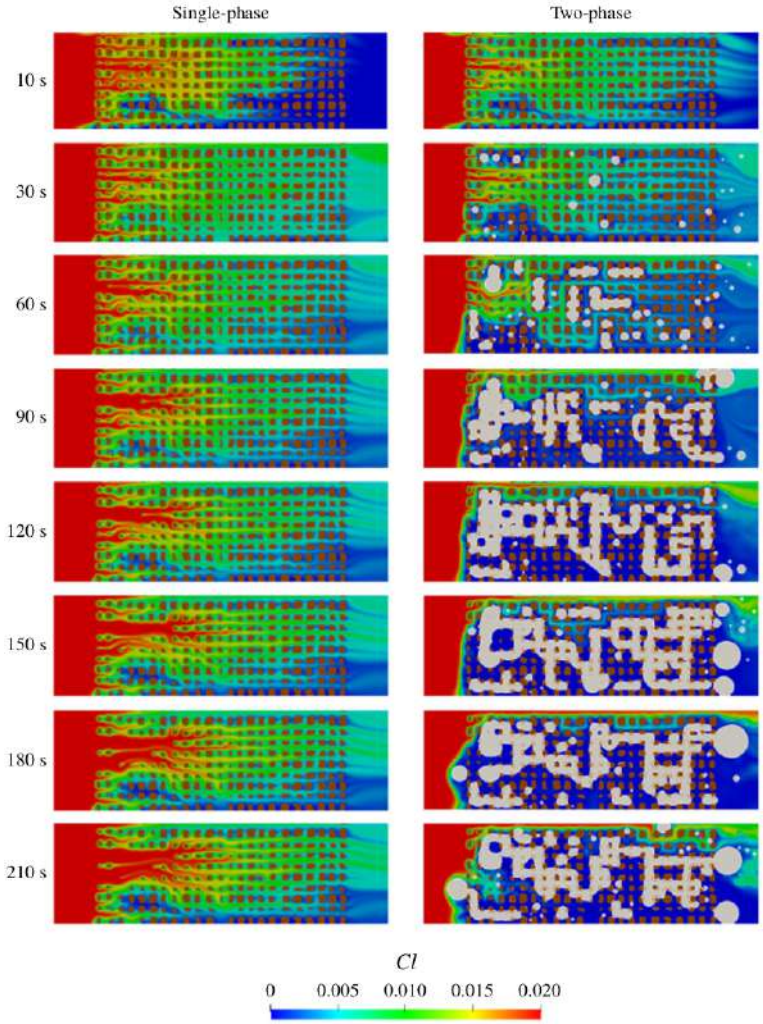


Figure 5.15: Evolution of a porous medium during the injection of acid. The colour map corresponds to the concentration of acid in the domain. Solid grains are depicted in brown. *(a)* Under single-phase flow conditions, the acid penetrates the domain and dissolves the solid grains producing wormholes. *(b)* The mineral dissolution generates CO_2 gas that occupies the pore space, limits the dissolution process and prevents the development of wormholes (from Soulaine et al. (2018)).

acid in the domain (see Fig. 5.15). These flow barriers prevent the emergence of wormholes and limit significantly the overall dissolution rate. This finding is of great interest in acid stimulation processes for which a gas phase can be produced (Prutton and Savage 1945; Thompson, Gdanski, et al. 1993). We indeed demonstrated that the presence of a second fluid phase under the same flow conditions may lead to very different dissolution regimes and that behaviour diagrams that characterize the dissolution pattern based on the Péclet and Damköhler numbers (Golfier et al. 2002) have to be complemented, at a minimum, by a third dimension that quantifies the solubility of the CO_2 in the aqueous phase.

Further improvements of the model include the consideration of gas compressibility and dynamic contact angle changes due to the evolution of the surface roughness during minerals dissolution.

5.3.3 Particulate transport

My current research (ANR Labex Voltaire2 TransCol project) intends to improve our knowledge on the role played by colloids (e.g. viruses, bacteria, microplastics, oil droplets, clay minerals) and fine particles in the soils and the subsurface. I am investigating the micro-dynamics of colloid aggregation and their stability, the attachment/detachment from the solid walls, and pores clogging mechanisms according to pH and salinity conditions. To that ends, I am looking at two complementary computational techniques: *i)* a CFD-DEM approach (Computational Fluid Dynamics - Discrete Elements Method) and *ii)* a micro-continuum approach. In the former, particles are tracked individually using a Lagrangian description that applies Newton's second law of motion to every particle while the flow is described by the local averaged Navier-Stokes equations (Tsuji et al. 1993). The approach implements a four-way coupling in the sense that the modeling account for the transport of the particles, the feedback on the flow due to the presence of the particles, and the mutual particle-particle interactions (O'Sullivan 2011). In the latter, particle transport, aggregation, and deposition are described with an Eulerian technique similar to the micro-continuum approach for reactive systems (Soulaine et al. 2021c). In both cases, the main challenge is to handle the strong coupling between hydro-electrochemical effects. This corresponds to the on-going PhD of Laurez Maya started in October 2021 (following a 6-months internship in the same topic).

5.4 Conclusion and perspectives

Computational microfluidics for geosciences offers an appealing framework to investigate the coupled processes in geological porous media. Modern approaches can simulate pore-scale events over a wide range of conditions including multiphase flow, microstructure evolution due to precipitation and dissolution, transport of particles, and biofilms growth. This modelling tool is essential to derive efficient continuum-scale models rooted in the elementary physical principles. In that regard, the computational microfluidics I am developing is based on multi-scale micro-continuum models, i.e. a unique set of partial differential equations are used both at the pore-scale and at the continuum-scale. The next Chapter reviews this modelling technique.

Chapter 6

Micro-continuum modelling: an hybrid-scale approach for solving coupled processes in porous media

I started to develop the concept of micro-continuum models in 2014 during my post-doc at Stanford University. They are versatile and powerful approaches for modelling coupled processes in two-scale systems. Initially oriented for modelling static single-phase flow in micro-CT images with sub-voxel porosity, the concept has been extended over the years to multi-phase flow, reactive transport, and poromechanics. This chapter is an introduction to the micro-continuum approach and a review of the state-of-the-art models developed by me, my co-workers, and the scientific community.

6.1 Introduction

Naturally occurring porous media involve a wide range of spatial scales. For example, the important contrast in pore-size distributions in fractured porous rocks comes from much larger characteristic lengths for the fractures than for the surrounding porous matrix. Therefore, the domain size required to reach an REV limits the use of pure pore-scale modelling. Hybrid-scale models have been proposed to describe systems that include multiple characteristic length-scales, for which some regions are described using pore-scale modelling (Navier-Stokes-based) while others are modelled with continuum approaches (Darcy-based).

Basically, two kinds of computational approaches exist to solve hybrid-scale problems. On the one hand, the domain decomposition technique solves different physics on separate domains – one for Darcy flow, another for Stokes (or Navier-Stokes) flow – linked together through appropriate boundary conditions (Molins et al. 2019). On the other hand, micro-continuum models use a single set of partial differential equations throughout the computational domain regardless of the content of a grid block (Soulaine and Tchelepi 2016; Steefel et al. 2015). In micro-continuum approaches, conditions at the interface between the two domains are included in the partial differential equations and automatically satisfied. In the latter approach, boundary conditions at the interface between the two regions are described as body source terms within the governing equation.

The micro-continuum approach is particularly well-suited to capture the dynamic

displacement of the interface between the porous and solid-free regions without involving complex re-meshing strategies. For example, micro-continuum models have been used successfully to simulate the formation and growth of wormholes in acidic environments (Golfier et al. 2002; Ormond and Ortoleva 2000; Soulaine and Tchelepi 2016). Hybrid-scale modelling is also a powerful tool in image-based simulations to account for microscale features that are not visible in the images because they are smaller than the imaging instrument resolution (Apourvari and Arns 2014; Arns et al. 2005; Scheibe et al. 2015; Soulaine et al. 2019, 2016).

Although the approach was initially dedicated to single-phase flow, we have made important efforts to extend the micro-continuum model to multi-phase flow (Carrillo et al. 2020; Soulaine et al. 2019, 2018).

This chapter is organized as follows. First (Section 6.2), we introduce the philosophy of the micro-continuum concept and its mathematical background. Second (Section 6.3), we present micro-continuum models for solving coupled processes including two-phase flow, heat transfer, hydro-geochemistry, and poromechanics. Then (Section 6.4), we present applications of micro-continuum models related to current research interests including Digital Rock Physics with sub-voxel porosity, and Reactive Transport Modelling of fractured systems. Finally (Section 6.5), we discuss open-challenges, and we close with a summary.

6.2 The micro-continuum approach

6.2.1 A description that depends on spatial resolution

The concept of micro-continuum is intimately related to the spatial resolution of the porous domain considered. The base element is the distribution of the fluid volume fraction, ϕ, in a cell of the computational grid. This volume fraction is therefore a cell porosity field. We have,

$$\phi = \begin{cases} 1, & \text{if the cell is occupied by fluids only,} \\ ]0,1[, & \text{if the cell contains a fluid-solid aggregate,} \\ 0, & \text{if the cell is occupied by solids only.} \end{cases} \tag{6.1}$$

Three cases of interest are illustrated in Figure 6.1. In the left panel, the medium porosity is fully resolved on the grid and ϕ has only two values, namely 0 and 1 (see Fig. 6.1a). This binary case corresponds to a pore-scale description with a sharp delineation between the fluid and the solid. The physics of flow is usually described by Navier-Stokes equations. In the right panel (see Fig. 6.1c), every cells of the grid contain a fluid-solid aggregate – i.e. an unresolved porous medium – and ϕ is always in the range $[0,1[$. There is always a minuscule amount of solid –even a tiny amount– in the grid blocks, and there is no cell containing fluid only. The physics of flow and transport is described by Darcy-based models. In between these two limit cases, there are intermediate situations for which some regions of the domain are fully resolved while others are unresolved (see middle panel on Fig. 6.1b).

The micro-continuum approach offers a unique framework –i.e. a unique set of partial differential equations– to describe flow and transport regardless of the spatial resolution. Importantly, micro-continuum models naturally handle hybrid-scale resolutions. They are also well-suited to simulate processes in which the solid mapping function evolves due to geochemistry or poromechanics.

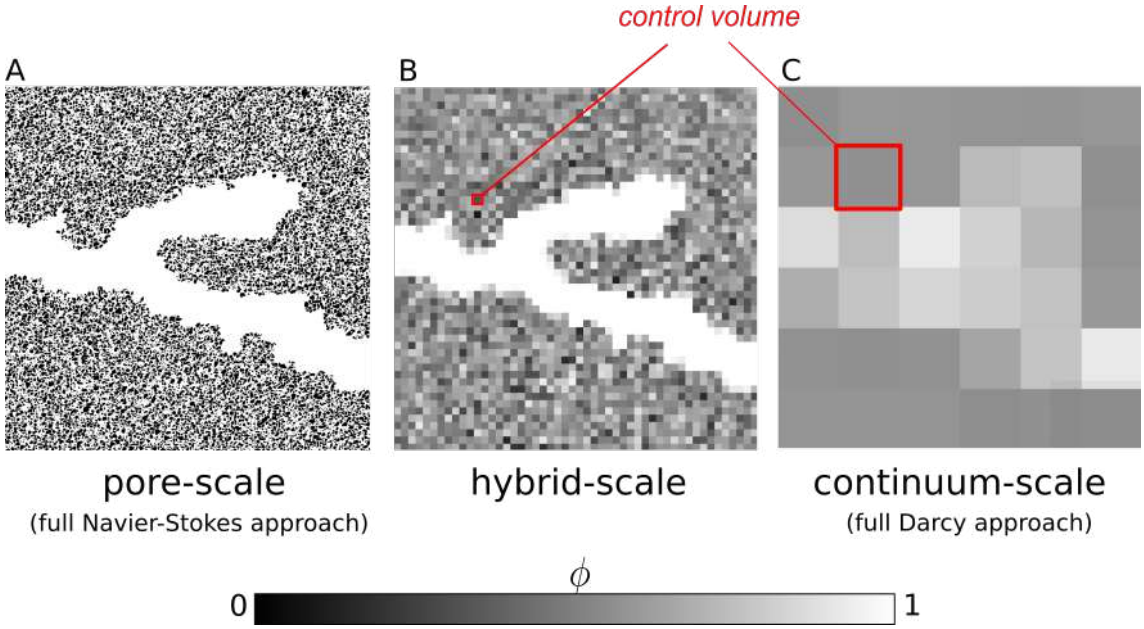


Figure 6.1: Schematic representations of a porous medium with two characteristic pore sizes depending on the scale of resolution: (a) full pore scale (Navier-Stokes), (b) intermediate or hybrid scale, and (c) full continuum scale (Darcy). The micro-continuum framework describes the physics of flow and transport using on a single set of equations resolved throughout the entire system regardless the spatial resolution.

6.2.2 The Darcy-Brinkman-Stokes equation

Micro-continuum equations are formed by cell-averaging the fundamental equations of continuum mechanics along with fluid-solid interfacial areas within the cell. Following the averaging procedure described in chapter 3, the integration of Navier-Stokes momentum equation over the cell volume, V_{cell}, gives (Whitaker 1986a,b),

$$\frac{1}{\phi}\left(\frac{\partial \rho \overline{\boldsymbol{v}}}{\partial t} + \nabla.\left(\frac{\rho}{\phi}\overline{\boldsymbol{v}}\overline{\boldsymbol{v}}\right)\right) = -\nabla\overline{p} + \rho\boldsymbol{g} + \nabla.\frac{\mu}{\phi}\left(\nabla\overline{\boldsymbol{v}} + \nabla^{t}\overline{\boldsymbol{v}}\right) - \mu k^{-1}\left(\overline{\boldsymbol{v}} - \overline{\boldsymbol{v}}_{s}\right) + \boldsymbol{F}, \quad (6.2)$$

where $\overline{\boldsymbol{v}} = \frac{1}{V_{\text{cell}}}\int_{V_f} \boldsymbol{v} dV$ and $\overline{p} = \frac{1}{V_f}\int_{V_f} p dV$ are the cell-averaged velocity and pressure, respectively (V_f being the volume occupied by fluids within V_{cell}). The left-hand side corresponds to inertia effects that are often neglected in subsurface flows. This momentum equation is an extension of the Darcy-Brinkman-Stokes (DBS) equation proposed by Brinkman (1947). The first and second terms of the right-hand side are the hydrodynamic driving forces, namely the pressure gradient and the gravitational acceleration. The third term describes the viscous dissipations due to the mutual friction of the fluid elements with each other. The fourth term is a drag force that describes the friction of the unresolved microstructure with the fluid. This flow resistance is characterized by the skin factor –also referred to as permeability– k^{-1} that is related to the geometry of the unresolved microstructure. $\overline{\boldsymbol{v}}_s$ is the velocity of the solid structure, e.g. because of poromechanical effects. If the structure is motionless, $\overline{\boldsymbol{v}}_s = \boldsymbol{0}$. Finally, the last term of the right-hand side is a body source term (e.g. surface tension forces, magnetic forces).

The extended DBS equation is generic and valid throughout the computational grid, regardless the cell content. Nonetheless, the form of the drag force and of

the body source term differs whether they describe the flow in a resolved or an unresolved region. If there is no solid in resolved regions there is no drag force, and the skin factor writes,

$$\mu k^{-1} = \begin{cases} 0, & \text{if the cell is occupied by fluids only,} \\ \mu k^{-1}, & \text{if the cell contains a fluid-solid aggregate.} \end{cases} \tag{6.3}$$

We will see in Section 6.2.5 that there exists constitutive laws such as Kozeny-Carman permeability-porosity relationship (Carman 1937; Kozeny 1927) for which the inverse of the permeability asymptotically tends toward zero if $\phi = 1$.

The body source force has different formulation in the resolved and unresolved regions. We simply write,

$$\boldsymbol{F} = \begin{cases} \boldsymbol{F}_{\text{resolved}}, & \text{if the cell is occupied by fluids only,} \\ \boldsymbol{F}_{\text{unresolved}}, & \text{if the cell contains a fluid-solid aggregate.} \end{cases} \tag{6.4}$$

For example, for immiscible two-phase flow, the surface tension forces are modelled by computing the interface of the curvature in the resolved region and the capillary pressure gradient in the unresolved region (Carrillo et al. 2020; Soulaine et al. 2019).

In practice, the DBS momentum equation is supplemented by, at a minimum, the fluid mass balance,

$$\frac{\partial \phi \rho}{\partial t} + \nabla . \left(\rho \overline{\boldsymbol{v}} \right) = 0. \tag{6.5}$$

Section 6.3 reviews the additional equations that need to be considered for modelling coupled processes including two-phase flow, heat transfer, reactive transport, and solid deformation.

6.2.3 Asymptotic behaviour

Asymptotic behaviour of the DBS equation

The most significant aspect of the DBS equation is its asymptotic behaviour across regions. On the one hand, in clear fluid regions, $\phi = 1$ and the drag force vanishes, and Eq (6.2) becomes the Navier-Stokes momentum balance,

$$\frac{\partial \rho \overline{\boldsymbol{v}}}{\partial t} + \nabla . \left(\rho \overline{\boldsymbol{v}} \overline{\boldsymbol{v}} \right) = -\nabla \overline{p} + \rho \boldsymbol{g} + \nabla . \mu \left(\nabla \overline{\boldsymbol{v}} + \nabla^t \overline{\boldsymbol{v}} \right) + \boldsymbol{F}_{\text{resolved}}, \text{ if } \phi = 1. \tag{6.6}$$

On the other hand, in porous regions, the drag force is dominant and the inertia effects and the viscous forces are negligible with respect to the other forces (Auriault 2009; Tam 1969; Whitaker 1986a). Therefore, Eq (6.2) tends asymptotically towards

$$0 = -\nabla \overline{p} + \rho \boldsymbol{g} - \mu k^{-1} \left(\overline{\boldsymbol{v}} - \overline{\boldsymbol{v}}_s \right) + \boldsymbol{F}_{\text{unresolved}}, \text{ if } \phi \in]0, 1[, \tag{6.7}$$

which is nothing else than Darcy's law.

Hence, thanks to this two limiting behaviours, the DBS equation, Eq. (6.2), is a unique partial differential equation that solves for flow in computational grids regardless of the spatial resolution (and therefore the content of a grid block). This feature is the cornerstone of the micro-continuum approach as it offers the possibilities for modelling processes in hybrid-scale domains that contain both resolved and unresolved regions.

Pore-scale modelling with immersed boundaries

If the porous matrix has sufficiently low permeability, fluid velocities in the porous domain are near zero and a no-slip condition is recovered at the interface between solid-free and porous regions (Angot et al. 1999; Khadra et al. 2000). This enables the use of micro-continuum simulations at the pore scale using a penalized approach, i.e., the solid phase is described as a low-permeability porous medium (Soulaine and Tchelepi 2016). Solving flow and transport through complex geometries using the penalized approach does not required complex grids – nor advanced softwares for meshing complex topology – as it relies on the mapping of the phase indicator function, ϕ, on Cartesian grids only.

Various techniques have been proposed –including some by the author– to describe the immersed boundary conditions at the fluid-solid interface for multiphase flows (Carrillo et al. 2020; Horgue et al. 2014; Soulaine et al. 2018) and chemical reactions at the solid surface (Molins et al. 2020; Soulaine et al. 2017b).

Using immersed boundaries on Cartesian grids is a great advantage for pore-scale simulations that involve the displacement of a fluid-solid interface due to geochemical processes such as dissolution and precipitation (Noiriel and Soulaine 2021; Soulaine et al. 2017b) or the mechanical deformation of the solid structure (Carrillo and Bourg 2019).

6.2.4 Interfacial conditions between resolved/unresolved regions

In hybrid-scale simulations, both solid-free and porous regions exist concomitantly in the computational grid (see Fig. 6.1b). Because the characteristic length size of the unresolved region may be several orders of magnitude lower than the one of the resolved porosity, there is no necessary continuity condition for the physical variables across the interface separating the two regions. Here, we present the state-of-the-art conditions at the interface between porous and solid-free domains for single-phase flow, two-phase flow, and species transport.

Single-phase flow condition

For single-phase flow, the continuity equation imposes a continuity of the normal component of the velocity field. Neale and Nader (1974) have demonstrated that the DBS equation captures the slip length induced by the continuity of the viscous stresses between the two regions as postulated by Beavers and Joseph (1967). This is illustrated in Figure 6.2.

Two-phase flow condition

For two-phase flow, the derivation of a comprehensive interfacial condition between the two regions still constitutes a major challenge to solve. Indeed, the discontinuity in porosity leads to a change in the form of the surface tension force. In our most recent work (Carrillo et al. 2020), we treat this discontinuity by assuming that the fluid-fluid interface of a droplet on a porous substrate forms a contact angle θ with the solid surface. The contact angle is an upscaled parameter that depends on various sub-grid scale properties including interfacial energies, surface roughness,

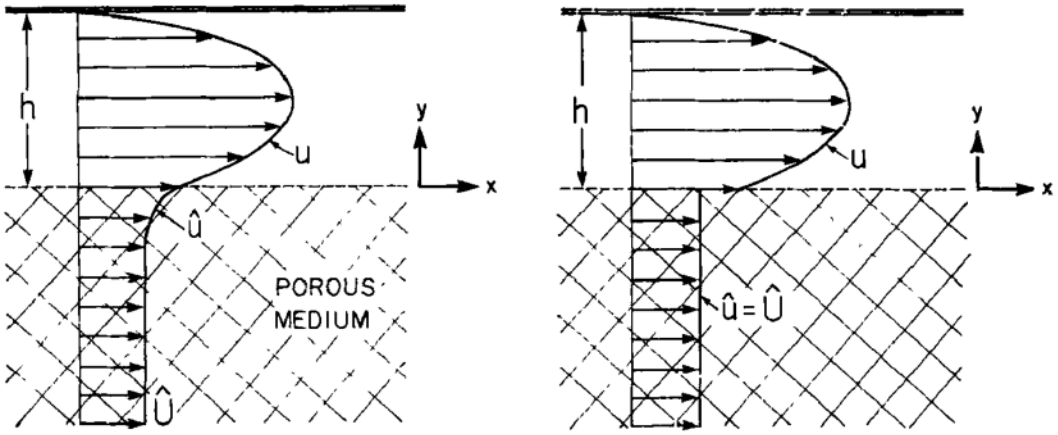


Figure 6.2: Interfacial conditions for single-phase flow between resolved and unresolved regions after Neale and Nader (1974). (Left) Actual velocity profile for coupled parallel flow within a channel and a bounding porous medium (Right) Velocity profile according to the slip-flow hypothesis of Beavers and Joseph (1967).

and the presence of thin precursor films (Cassie and Baxter 1944; Meakin and Tartakovsky 2009; Wenzel 1936). In our current numerical model, the contact angle is imposed by locally modifying the orientation of the fluid-fluid interface relative to the solid surface (Horgue et al. 2014; Soulaine et al. 2019, 2018). Further works are still needed to improve this interfacial condition.

Non-equilibrium solute transport condition

Modelling solute transport across the interface between the resolved and the unresolved regions also remains a challenge because of the scale separation that exists between both regions. The situation becomes even more complex if the interface itself is a membrane whose thickness is not represented in the computational domain, but whose effects on mass transfer can not be neglected because it implies a discontinuity on the concentration profile. In all cases, there is a continuity of the mass flux, F, in the direction normal to the interface. A generic formulation of the thermodynamic condition at the interface writes,

$$F = \alpha \left(C_{\text{res.}} - H C_{\text{unres.}} \right), \tag{6.8}$$

where $C_{\text{res.}}$ and $C_{\text{unres.}}$ are, respectively, the species concentrations on the resolved and unresolved sides of the interface, H is a partitioning parameter that characterizes the concentration discontinuity due to the membrane at the equilibrium, and the kinetic parameter α (in m/s) is a physical property of the interface that controls the delay to reach the equilibrium. When there is no more mass transfer, $F = 0$ and $C_{\text{res.}} = H C_{\text{unres.}}$, and the system is at equilibrium. The non-equilibrium condition has two asymptotic regimes whether α is large or small, i.e. the time scale to reach the equilibrium is very high or very low. On the one hand, the equilibrium condition, $C_{\text{res.}} = H C_{\text{unres.}}$, is recovered if $\alpha \to \infty$. The latter condition means that the time delay to reach the equilibrium is so small that the equilibrium is reached instantaneously. On the other hand, if $\alpha \to 0$, then $F \to 0$ and the transport in the two regions is uncoupled. The interface behaves as an inert impervious barrier.

We are currently developing a generic non-equilibrium model for species transport across interfaces. The numerical model is formulated in terms of a single-field partial differential equation along the same lines of the DBS model –i.e. an unique equation valid throughout the computational domain regardless the cell content (resolved or unresolved)– and automatically handles the non-equilibrium condition, Eq. (6.8), at the interface.

6.2.5 Sub-grid models

In the micro-continuum framework, the physics of flow and transport in the unresolved regions is filtered and modelled through the concept of sub-grid models. These describe the processes occurring below the grid-size. Because they are not resolved, these processes have to be modelled through constitutive laws that are provided to the micro-continuum model. In practice, sub-grid models rely on the classic concept used for modelling flow and transport in porous media.

For example, the parameter k, in Eq. (6.7) is a cell permeability that characterizes the sub-grid microstructure. There is no unique formulation for the sub-grid permeability but a large variety of models that depend on the phenomena involved. It can be constant for low flow rates or varying with the velocity field for fast flow rates due to the Forchheimer correction to Darcy's law (Forchheimer 1901; Soulaine and Quintard 2014). If the cell contains several fluid phases, then the permeability depends on the fluid saturation using the classic relative permeability concepts (Carrillo et al. 2020; Soulaine et al. 2019). For submicron pore throats, nanoscale effects are no longer negligible and the permeability is modelled using Klinkenberg's law (Klinkenberg 1941). This sub-grid model allows us to simulate shale gas recovery from nanoporous kerogen (Soulaine et al. 2019).

The body source term, $\boldsymbol{F}$, has often a different formulation in the resolved and unresolved regions. For example, the surface tension force is described in the resolved domain by computing the curvature of the fluid-fluid interface (Brackbill et al. 1992) and in the unresolved region using the concept of capillary pressure curves including Brooks and Corey (1964) and van Genuchten (1980) models.

For solute transport in the unresolved regions, the concept of tortuosity and hydrodynamic dispersion apply. Specific surface area in a grid-block is a key sub-grid model in case of geochemical reactions.

6.3 Micro-continuum models for coupled processes

Micro-continuum models have the capacity for modelling coupled processes. This is achieved by combining the Darcy-Brinkman-Stokes momentum equation introduced in the previous section with additional physics. In the following section, we discuss coupled micro-continuum models including two-phase flow, heat transfer, geochemistry, and poromechanics.

6.3.1 Two-phase flow micro-continuum models

Micro-continuum models for two-phase flow involve the evolution of the wetting phase saturation, S_w. The partial differential equation for the saturation reads (Carrillo et al. 2020; Soulaine et al. 2019, 2018),

$$\frac{\partial \phi S_w}{\partial t} + \nabla . \left(\overline{\boldsymbol{v}} S_w \right) + \nabla . \left(\phi S_w \left(1 - S_w \right) \overline{\boldsymbol{v}}_r \right) = 0, \tag{6.9}$$

where $\overline{\boldsymbol{v}}_r$ is multi-scale parameter the describes the average relative velocity between the two fluid phases in a grid block. This term has different forms in the resolved and in the unresolved regions. Carrillo et al. (2020) derived a model for $\overline{\boldsymbol{v}}_r$ in which the saturation equation tends on the one hand to the evolution of the Volume-of-Fluid phase indicator function in resolved region, and on the other hand to the classic saturation equation used in porous media modelling.

In two-fluid systems, the permeability, k, in Eq. 6.2 relies on relative permeabilities that depend on S_w (Carrillo et al. 2020; Soulaine et al. 2019). The surface tension effects between the two immiscible fluids are modelled as a body source term $\boldsymbol{F}$ in the DBS momentum equation. In the resolved region, $\boldsymbol{F}$ corresponds to the Continuum Surface Force computed from the curvature of the interface (Brackbill et al. 1992). In the unresolved region, $\boldsymbol{F}$ depends on the gradient of the capillary pressure curves (Carrillo et al. 2020). Relative permeabilities and capillary pressure curves are sub-grid models classically described using van Genuchten (1980) and Brooks and Corey (1964) formula.

6.3.2 Heat transfer with micro-continuum approaches

Micro-continuum models for heat transfer rely on a thermal equilibrium model in the unresolved regions, i.e. a single-field temperature, T, – corresponding to a mixture of the fluid and solid temperatures – describes the heat evolution in the system (Soulaine and Tchelepi 2016). The governing equation writes,

$$\left(\phi \left(\rho_f C p_f \right) + \left(1 - \phi \right) \left(\rho_s C p_s \right) \right) \frac{\partial T}{\partial t} + \left(\rho_f C p_f \right) \overline{\boldsymbol{v}} . \nabla T = \nabla . \left(\lambda \nabla T \right) + Q_T, \tag{6.10}$$

where $\left(\rho_f C p_f \right)$ and $\left(\rho_s C p_s \right)$ are the fluid and solid heat capacities, λ is the single-field thermal conductivity, and Q_T is a heat source/sink term (e.g. enthalpy of reaction). The thermal conductivity, λ, is a mixture of the fluid and solid conductivities, λ_f and λ_s. Maes and Menke (2021) use an harmonic average, $\lambda = \frac{\lambda_f \lambda_s}{\phi \lambda_s + (1-\phi) \lambda_f}$.

In the resolved regions, Eq. 6.10 tends to the fluid temperature equation mainly driven by advection. In low-porosity low-permeability unresolved regions, Eq. 6.10 tends to the solid temperature equation driven by thermal conductivity.

The thermal equilibrium base hypothesis of the heat transfer micro-continuum model is limiting in the case of advection-dominated regimes in unresolved regions.

6.3.3 Modelling hydro-bio-geochemical processes

The main idea of micro-continuum models for hydro-geochemical processes is to describe the evolution of the volume fraction of N_s different minerals, $Y_{s,i}\left(x, y, z, t\right)$, in the computational grid. This is achieved using

$$\rho_{s,i} \frac{\partial Y_{s,i}}{\partial t} = -\dot{m}_{s,i} \text{ for } i = 1, .., N_s, \tag{6.11}$$

where $\rho_{s,i}$ is the mineral density and $\dot{m}_{s,i}$ is the rate of solid change. The latter is calculated from the aqueous species transport using the mass balance equations,

$$\frac{\partial \phi C_j}{\partial t} + \nabla.\left(\overline{\boldsymbol{v}} C_j\right) - \nabla.\left(\phi \mathsf{D}_j^*.\nabla C_j\right) = \sum_{i=1}^{N_s} \dot{m}_{f_j,i} \tag{6.12}$$

where C_j is the concentration of species j, D_j^* is the dispersion tensor including molecular diffusion, tortuosity and hydrodynamic dispersion, and $\dot{m}_{f_j,i}$ describes chemical reactions at the fluid-rock interface. Surface reaction are often modelled as kinetic laws proportional to the mineral surface area per unit of volume. The latter is assessed with different sub-grid models whether the surface reaction occurs within the unresolved region or at the interface between the resolved and unresolved domains. In the first case, sub-grid models estimate the accessible reactive surface area as a function of the solid volume fraction and local flow rate (Noiriel et al. 2009; Soulaine et al. 2017b). On the other case, the surface area is computed directly from the delineation between the two regions using the Volume-of-Solid approach, i.e. by computing gradient of the solid volume fraction (Soulaine et al. 2017b).

The evolution of the porosity field is obtained using:

$$\phi = 1 - \sum_{i=1}^{N_s} Y_{s,i}. \tag{6.13}$$

The variation of the porosity field informs the displacement of the resolved-unresolved interface by updating the local permeability in Eq. 6.2.

More details about multi-scale reactive transport modelling including the coupling with geochemical packages is found in Soulaine et al. (2021c).

6.3.4 Solid deformation modelling: Darcy-Brinkman-Biot

The Darcy-Brinkman-Biot model proposed by Carrillo and Bourg (2019) describes hydro-mechanical coupling in two-scale deformable porous media. In addition to the micro-continuum fluid momentum and continuity equations, their model considers a mass balance and a momentum equation for the solid phase as well. For plastic and elastic solids, it writes,

$$\frac{\partial \phi_s \rho_s}{\partial t} + \nabla.\left(\phi_s \rho_s \overline{\boldsymbol{v}}_s\right) = 0, \tag{6.14}$$

and,

$$-\nabla.\sigma = \phi_s \nabla.\tau + \phi \mu k^{-1}\left(\overline{\boldsymbol{v}} - \overline{\boldsymbol{v}}_s\right), \tag{6.15}$$

where ρ_s is the solid density, $\overline{\boldsymbol{v}}_s$ is the solid velocity, τ is the Terzaghi stress tensor defining the effective stress due to both fluid pressure and confining pressure, and σ is the plastic (or elastic) stress tensor. Note that the last term is a mutual momentum transfer due to the viscous resistance (i.e. drag) of the fluid on the solid surface.

According to poromechanics theory, the Terzaghi stress tensor writes,

$$\tau = \sigma_{\text{conf}} - \overline{p} b \mathsf{I}, \tag{6.16}$$

where σ_{conf} is the confining pressure, b is the Biot coefficient, and $\overline{p}$ is the fluid pressure. The Biot coefficient describes how the bulk volume changes relative to

the fluid pressure at a fixed stress. For modelling clay swelling, Carrillo and Bourg (2019) also add a swelling pressure.

For compressible plastic solid, the stress tensor is,

$$\sigma = \phi_s \mu_s^{eff} \left(\nabla \overline{\boldsymbol{v}}_s + \nabla^t \overline{\boldsymbol{v}}_s - \frac{2}{3} \left(\nabla . \overline{\boldsymbol{v}}_s \right) \mathsf{I} \right), \tag{6.17}$$

where the effective solid viscosity μ_s^{eff} can be described using various non-Newtonian plastic viscosity models.

Within the Darcy-Brinkman-Biot framework, recently Carrillo and Bourg (2021) propose a model for deformable porous materials in two-fluid systems.

6.4 Applications in geosciences

In this section, cutting-edge micro-continuum models are used to simulate porous media processes. First, we highlight the ability of micro-continuum models to account for subvoxel porosity in image-based simulations. Then, we review recent work on multi-scale reactive transport modelling.

6.4.1 Digital Rock Physics and micro-continuum models

Digital Rock Physics – a new scientific discipline that emerged from the development of High-Performing Computing combined with high-resolution imaging – aims at the evaluation of rock properties based on physical simulations (Andrä et al. 2013a,b; Blunt et al. 2013). For example, by volume averaging the velocity solution of a Stokes flow in a porous rock sample imaged with micro-CT one can get an estimate of the absolute permeability. Micro-continuum models push the limit of current Digital Rock Physics capabilities by considering sub-voxel porosity and coupled processes.

Digital Rock Physics with subvoxel porosity

Sub-resolution porosity (SRP) is an ubiquitous, yet often ignored, feature in Digital Rock Physics. It embodies the trade-off between image resolution and field-of-view, and it is a direct result of choosing an imaging resolution that is larger than the smallest pores in a heterogeneous rock sample. The micro-continuum approach presents an alternative route to simulating dynamic flow processes in systems with SRP. In this context, flow and transport in unresolved pores are modelled using porous media concepts.

Micro-continuum models are fairly flexible to describe digital rock systems including two characteristic length scales. They can account for the impact of SRP on the estimation of the rock sample absolute permeability (Apourvari and Arns 2014; Kang et al. 2019; Knackstedt et al. 2006; Scheibe et al. 2015; Singh 2019; Soulaine and Tchelepi 2016). The sub-grid permeability that quantifies the flow resistance of the microstructure that is not visible in the image is described using the only information within reach. Soulaine and Tchelepi (2016) uses a Kozeny-Carman law including the cell porosity and the imaging instrument resolution. They observe that SRP in a Berea sandstone imaged with a $3.16\,\mu\text{m}^3$/voxel resolution have a disproportionately large impact on permeability – up to a factor 2 for only 2% of voxels

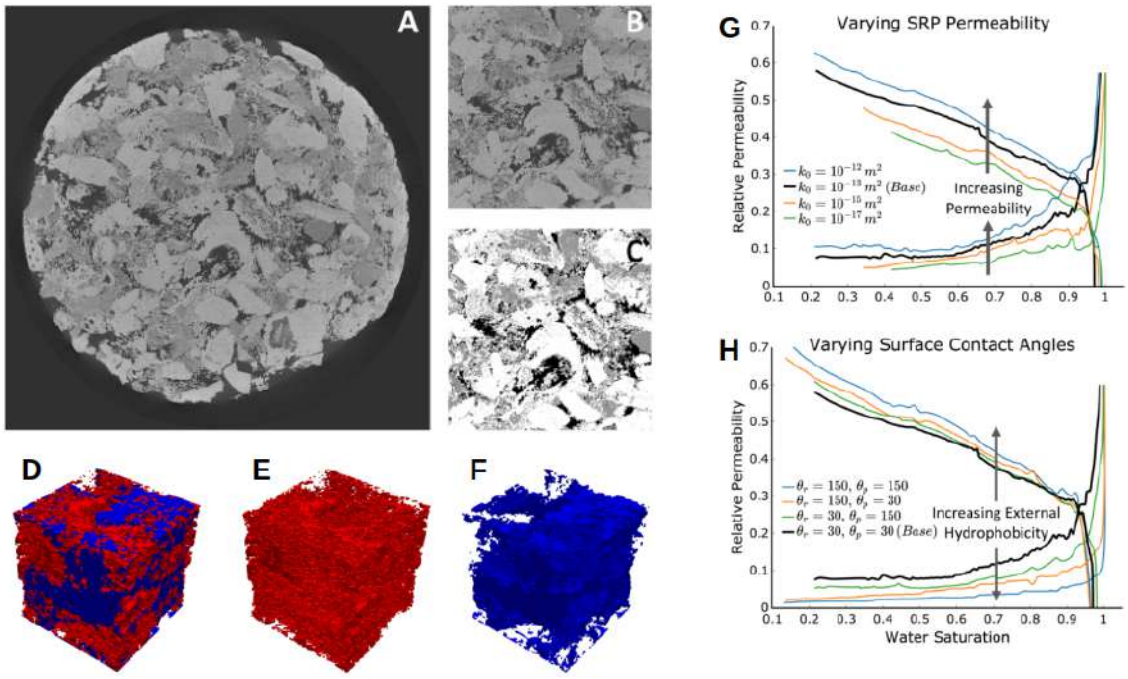


Figure 6.3: The impact of sub-voxel on rock properties in Digital Rock Physics, obtained from Carrillo et al. (2021). (A) Full 2-D view of the XCT scan of Estaillades carbonate rock sample by Bultreys (2016), which is 7 mm in diameter with a resolution of 3.1 μm per voxel. Black is open pore space, dark grey corresponds to domains that contain SRP, and the lightest color is solid calcite. (B) 500×500 voxel cropped sample. (C) Corresponding segmented image. (D) Spatial distribution of the SRP (red), pore space (blue), and solid rock (transparent) within the extracted 3-D rock representation ($200 \times 200 \times 200$ cells). (E) The corresponding SRP, which accounts for 21% of the voxels. (F) The associated open pore space, which accounts for 40% of the voxels. (G) Sensitivity of drainage and imbibition relative permeability curves to SRP absolute permeability, from $k_0 = 10^{-17}$ to 10^{-12} m^2. (H) Sensitivity of drainage and imbibition relative permeability curves to the external wettability of rock and SRP domains, from water-wetting ($\theta_r = 30°,\ \theta_p = 30°$), to mixed-wetting ($\theta_r = 150,\ \theta_p = 30°$ and $\theta_r = 30°,\ \theta_p = 150°$), to oil-wetting ($\theta_r = 150°,\ \theta_p = 150°$).

containing SRP if they are neglected – implying that SRP forms key percolation pathways for fluid flow.

Using the newly developed micro-continuum approach for multiple fluids in multiscale porous media (Carrillo et al. 2020; Soulaine et al. 2018), Carrillo et al. (2021) investigated the effects of micro-CT sub-resolution porosity on a rock's absolute permeability, relative permeability, residual saturations, and fluid breakthrough times (see Figure 6.3). Sub-grid models for two-phase flow processes in SRP correspond to the relative permeability and capillary pressure curve of van Genuchten (1980). Their results quantify how these four properties react to changes in the porosity, permeability, and wettability of the SRP. One notable finding is that SRP can function as a persistent connector between otherwise-isolated fluid clusters during multiphase flow, even at low saturations. To the best of our knowledge, this is the first two-phase flow model and study to take into account SRP without having to rely on a quasi-static assumption or simplified pore-network models. Overall, the

study of Carrillo et al. (2021) confirms previous evidence that the influence of SRP cannot be disregarded without incurring significant errors in numerical predictions or experimental analyses of multiphase flow in heterogeneous porous media. The two-phase micro-continuum DRP will lead the way towards a greater understanding of multiscale rock physics and the development of more accurate and predictive upscaled permeability models.

Digital Rock Physics for characterizing heat transfer

Recently, Maes and Menke (2021) modelled conjugate heat transfer during injection of cold water into a micro-CT image of Bentheimer sandstone which is analogous to a geothermal reservoir using a micro-continuum-based approach (Fig. 6.4). They considered the solid grains as low-permeability low-porosity porous media in which the temperature evolution depends only on thermal diffusion. They then performed simulations with various fluid properties at various Reynolds numbers in order to investigate the evolution of the Nusselt numbers which describe the heat exchange between solid and fluid. Finally, they obtained different trends for the heat exchange coefficient whether the system was in the conduction-dominated regime or in the convection-dominated regime.

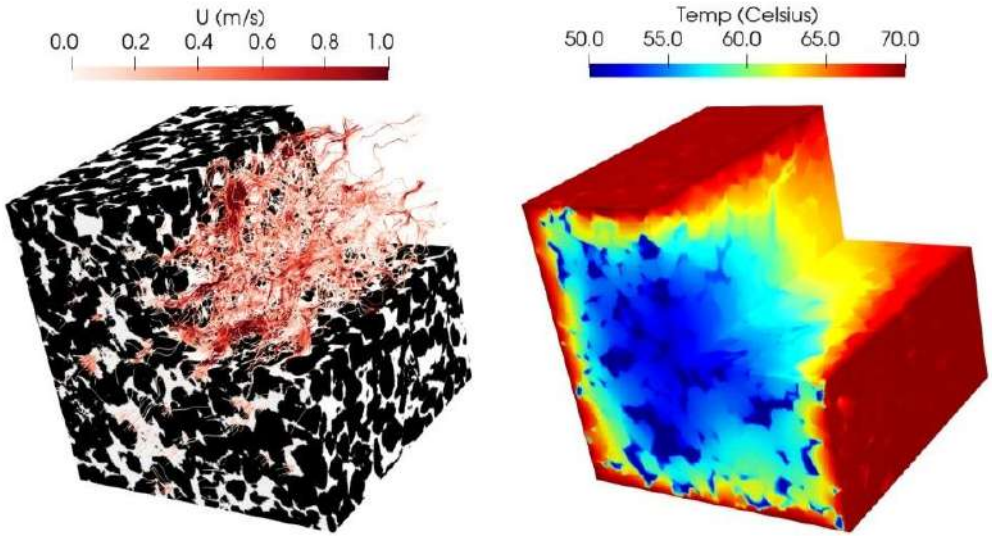


Figure 6.4: Heat transfer using Digital Rock Physics after Maes and Menke (2021). Computational domain is a 400^3 voxel micro-CT image of Bentheimer sandstone with a resolution of 5 microns. Fluid is injected from the left boundary at constant velocity and constant temperature $T = 50\,°\text{C}$, and exits the domain at the right boundary. The top, front, bottom and back boundaries have a no-flow condition and a constant temperature $T = 70\,°\text{C}$.

6.4.2 Reactive Transport Modelling

Over the last decades, reactive transport modelling has become an essential tool for the study of subsurface processes involving flow, transport and geochemical reactions (Steefel et al. 2005). This discipline arises from the junction of two scientific communities, namely geochemistry and transport in porous media. They consist of

computational models that describe the coupled physical, chemical, and biological processes that interact with each other over a broad range of spatial and temporal scales. Because they have the ability to displace fluid-rock interfaces on Cartesian grids, micro-continuum models have demonstrated great capacities to model geochemical processes at the different scales of interest (Soulaine et al. 2021c, 2017b, 2018; Soulaine and Tchelepi 2016; Steefel et al. 2015).

In the following section, we show recent applications of micro-continuum-based reactive transport modelling for simulating hydro-geochemical processes.

Pore-scale modelling of dissolution processes

The[1] evolution of surface roughness in acidic environments has important impacts on many transport processes. Notable consequences include changes in absolute permeability (Pilotti et al. 2002), alteration of wettability (Wan et al. 2014), and reduction of the large-scale reaction rates caused by the presence of eddies within the roughness cavities (Deng et al. 2018). Here, we investigate the evolution of surface roughness in diffusion-dominated regimes using high-resolution pore-scale simulations.

We consider a two-dimensional $512 \times 249\,\mu\text{m}^2$ domain that corresponds to the vicinity of the fluid-rock interface. The interface profile is derived from statistical properties (roughness and standard deviation of the surface elevation) calculated from the initial 512×512 surface topography presented in Noiriel and Soulaine (2021). The solid phase is mapped onto the computational grid using the mineral volume fraction, Y_s (see Figure 6.5). Volume fractions $Y_s = 1$ denotes cells that contain the solid phase while $Y_s = 0$ corresponds to cells occupied by water. Cells with intermediate values, $0 < Y_s < 1$, contain the fluid-rock interface. The initial roughness is $704\,\mu\text{m}$ long. In this illustration, there is no flow. The chemical species are transported by diffusion ($D_i = 10^{-9}\,\text{m}^2/\text{s}$) from the bulk to the mineral surface where geochemical reactions lead to a retreat of the fluid-rock interface. Initially, the concentration of acid in the domain is zero. A fixed concentration value, $C_{H^+} = 10^{-2}\,\text{mol}/\text{m}^3$, is applied at the top boundary to describe a constant supply of HCl far from fluid-rock interface. The lateral boundary conditions are periodic, and the bottom boundary is impermeable. The reactive condition is applied at the fluid-rock interface using immersed boundary conditions. This is achieved using the Volume of Solid approach for which the surface of the fluid-rock interface within a grid block, A_e, is estimated from the gradient of the mineral distribution (Soulaine et al. 2017b). The evolution of the surface roughness is simulated during 500 hours for Damköhler numbers $Da = \frac{k_{H+}}{AeD_i}$ ranging from 10^{-1} to 10^3 by varying the constant of reaction.

Results are presented in Figure 6.5. For low Damköhler numbers, i.e., $Da < 1$, the characteristic time scale of species transport in the vicinity of the fluid-rock interface is much lower than the typical time of reaction processes at the mineral surface. Therefore, the species concentration is uniform at the solid surface and the interface recedes uniformly at the same velocity. During the mineral dissolution, we observe a translation of the interface towards the bottom of the domain with quasi conservation of the surface roughness. For larger Damköhler numbers, i.e. $Da > 1$, the surface reaction is faster than the travel time of the chemical species to the fluid-mineral interface. As the chemical species have a lower distance to travel

[1] This part has been published in Noiriel and Soulaine (2021).

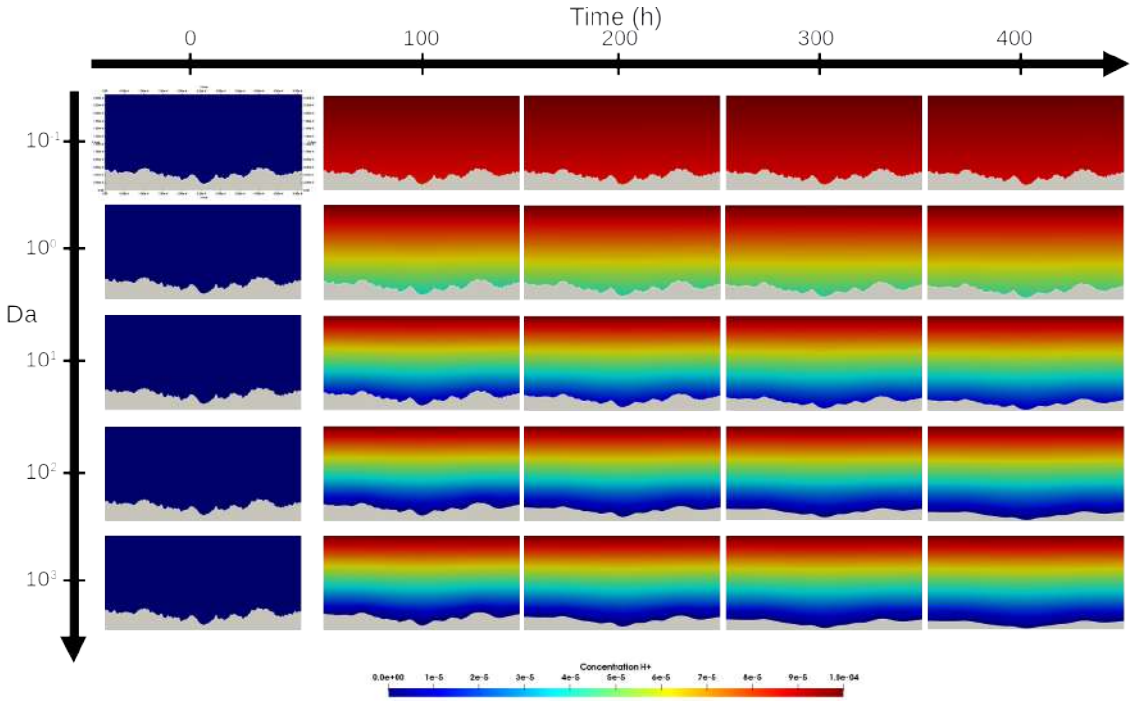


Figure 6.5: Evolution of the surface roughness for different Damköhler (Da) numbers in a diffusion-dominated transport regime. For $Da < 1$, the distribution of the chemical species at the fluid-mineral interface is uniform and the solid dissolves conserving the surface roughness. For $Da > 1$, the peaks are dissolved before the topographical lows causing a flattening of the surface roughness. From Noiriel and Soulaine (2021).

to reach the peaks compared with the hollow regions of the roughness, there is a non-uniform distribution of reactants at the interface. These local concentration gradients favor the dissolution of the peaks which smooth down the roughness. The phenomenon is more pronounced for $Da > 10$ as illustrated in Figure 6.5. The flattening of the surface roughness induced by mineral dissolution could explain, at least partially, the change of contact angle measured experimentally when a droplet of supercritical CO_2 surrounded by brine is deposited on the surface of a reactive mineral (Wan et al. 2014). The new possibilities offered by pore-scale simulators with evolving fluid-rock interface will be used to investigate the impact of surface roughness alteration on coupled hydro-geochemical processes.

Fracture-Matrix interaction

Image-based[2] simulations offer an appealing framework to investigate fracture-matrix interactions in reactive environments. The large contrast of characteristic length scales involved in fractured porous media – the typical pore sizes of the rock matrix and of the fracture aperture differ by several orders of magnitude – limits the use of very high-resolution images that fully resolve both the fracture and the matrix microstructure. Nevertheless, the presence of the surrounding porous matrix can impact the evolution of the fracture aperture by the development of a weathered zone at the vicinity of the fracture-matrix interface and must be included in the

[2]This part has been published in Noiriel and Soulaine (2021).

modelling. Here, we use pore-scale simulations to compare the evolution of a fracture geometry during the injection of acidic brine in two cases: (i) the rock matrix is impermeable and (ii) the rock matrix is porous.

The fracture geometry corresponds to the 3D micro-tomography images obtained in Noiriel et al. 2007. For the simulations, we consider that the reacting matrix contains only calcite. The mineral distribution is mapped onto a $25 \times 100 \times 165$ Cartesian grid directly derived from the segmented volume of interest. Two 75 μm long manifolds are added at the inlet and outlet of the computational domain to facilitate injection conditions. An aqueous solution at pH=2 is injected for 100 hours. A pressure difference between the inlet and the outlet, $\Delta P = 0.6\,\text{Pa}$, is applied throughout the injection of solute in the system. The resulting flow rates are in the order of $100\,\text{cm}^3/\text{h}$ as reported in Noiriel et al. 2007.

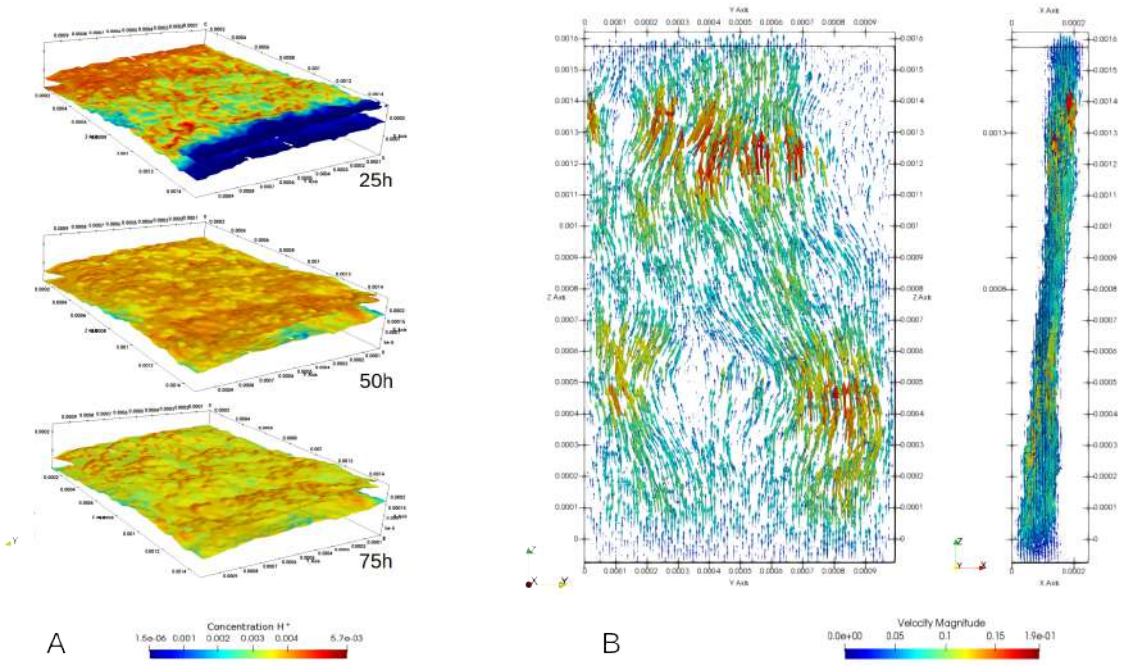


Figure 6.6: Fracture response to the injection of HCl. (a) Evolution of the fracture aperture after 25, 50 and 75 hours. (b) Velocity vectors within the fracture. The non-uniform aperture of the fracture results in a heterogeneous velocity pattern with faster and slower flow regions (from Noiriel and Soulaine (2021)).

In both cases, the micro-continuum approach is used for solving Stokes equations in the fracture and Darcy's law in the matrix. A Kozeny-Carman law allows switching automatically from Stokes (in the fracture) to Darcy (in the matrix) whenever the local porosity, ϕ, is smaller than 1. The fracture-matrix interface is displaced according to chemical reactions that modify the porosity, which subsequently updates the local permeability field and then the flow profile. The impermeable and porous matrix cases differ, on the one hand, by the initial permeability and porosity values, and on the other hand, by the model used to compute the mineral-fluid surface area, A_e, in each grid block. In the first case, a fictitious low-porosity low-permeability matrix is used to describe impermeable boundaries (initially, $\phi_0 = 0.001$ and $k_0 = 10^{-20}\,\text{m}^2$). Porosity value between 0 and 1 ($0 < \phi < 1$) denotes computational cells that contain the sharp fluid-rock interface. The Volume of Solid approach (Soulaine et al.

2017b) uses the gradient of the mineral volume fraction, Y_s, to compute the surface area from the fracture geometry and apply surface reaction at the rock-fluid interface. In the porous matrix case, the matrix porosity and permeability are initially $\phi_0 = 0.2$ and $k_0 = 10^{-13}\,\mathrm{m}^2$, respectively. The displacement of a porous-fluid interface is a two-scale problem and accurate modelling depends on the constitutive law used to describe the mineral surface area within a computational grid block. For the fracture-matrix interface, it seems relevant to assume that the species transport from the fracture to the host matrix is dominated by diffusion. Therefore, in the simulations, the surface area within a computational grid block evolves according to a power-law, $\frac{A_e}{A_0} = \left(\frac{Y_s}{Y_{s,0}}\right)^n$, where the initial surface area of the host matrix is $A_0 = 6500\,\mathrm{m}^2/\mathrm{m}^3$ and $n = 2/3$ is a geometric parameter (Noiriel et al. 2009).

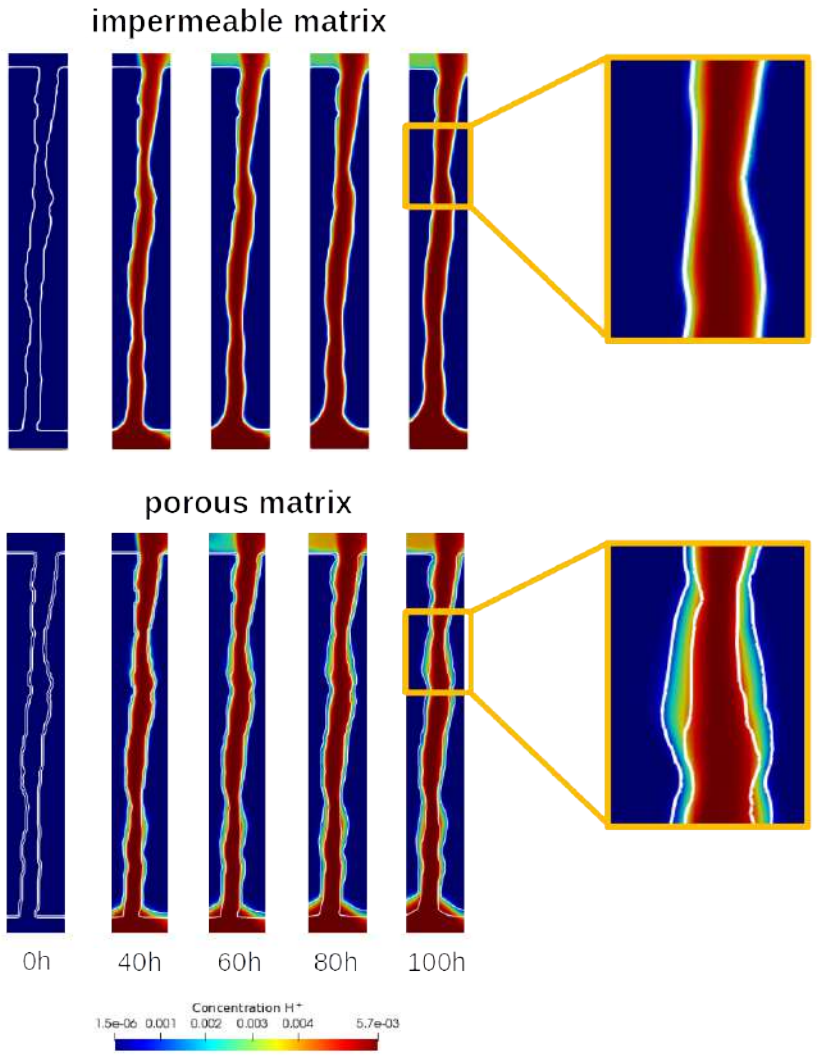


Figure 6.7: Two-dimensional cross-section of the fracture aperture, with the host matrix either impermeable or porous. The white lines correspond to porosity isocontours ($\phi = 0.5$ for the impermeable case, and $\phi = 0.4$ and $\phi = 0.25$ for the porous case). Note the development of an altered layer due to the lateral penetration and reaction of the acidic solution into the host matrix (from Noiriel and Soulaine (2021)).

The fracture aperture evolution results from a complex interplay between ad-

vection, diffusion, and surface reaction. As illustrated in Fig. 6.6b, the velocity profile within the fracture is heterogeneous with fast flow regions and slower flow regions due to the topology of the fracture boundaries. In advection-dominated transport regimes, the faster flow pathways preferentially drive the reactant which leads to a non-uniform dissolution of the fracture surface, a phenomenon known as channeling (Starchenko and Ladd 2018). Using impermeable boundaries, the mass of solid mineral decreases by 10 % between the initial and final stage of the acid injection (see Figure 6.6a and Figure 6.7a). As the fracture aperture increases, the system permeability – estimated to be initially $10^{-10}\,\mathrm{m}^2$ – increases by 150 % after 100 hours of HCl injection. The presence of a porous host matrix strongly impacts the evolution of the fracture aperture and of the sample permeability. We obtain a decrease of the mass of calcite by 6 % as well as a permeability increase of 50 %. Unlike the evolution of the fracture aperture within an impermeable host matrix that is always sharp, the porous-fluid interface can be diffused forming a weathered zone as observed in Figure 6.7b. The development of this altered layer depends on the penetration length of chemical species into the host matrix that varies according to the interplay between advection, diffusion, and surface reaction (Deng et al. 2016, 2017). The simulations presented in this part highlight the capabilities of the micro-continuum approach to capture the development of altered zones. Nevertheless, accurate modelling of the fracture-matrix interface using hybrid-scale models needs a proper description of the matrix properties evolution during geochemical processes. According to matrix mineralogy, other constitutive laws can be more relevant. For example, in the case of the dissolution of an aggregate of mineral particles, the sugar-lump model (Noiriel et al. 2009) increases the surface area per unit of volume while the power-law used in the present simulations decreases the specific surface area. Further investigations will focus on the characterisation of the altered layer thickness according to the Péclet and the Damköhler numbers.

Wormholes formation in acidic environments

The micro-continuum approach is particularly well-suited to simulate the formation and growth of wormholes in acidic environments without involving complex re-meshing strategies (Golfier et al. 2002; Ormond and Ortoleva 2000; Soulaine and Tchelepi 2016). The modelling of such structures is particularly challenging, as they are intrinsically multi-scale and dynamic. They constitute a major procedure in the process of acidizing wells to enhance oil productivity.

Figure 6.8 presents the emergence of wormholes in a 10 cm long carbonate core in which hydrochloric acid is injected at a constant flow rate. For improving the visualization of the results, we show the cells that contain fluid only, i.e. the channel formed by the dissolution of the solid minerals. At early times, instabilities begin to develop when the reactant preferentially penetrates the regions with the biggest pores (the cells with the highest porosity and permeability) and erodes the walls to form the flow channels. When a finger is slightly longer than its surrounding neighbours, the pressure drop in this finger is smaller than in the surrounding fingers, which favors a preferential flow through the finger and reduces the growth of the other surrounding smaller fingers. The process continues until only one dominant wormhole takes over. After the breakthrough of this wormhole, the dissolution dynamic changes. The acid mostly flows through the wormhole, and since the characteristic time of the reaction is much larger than the transport times, the dissolution

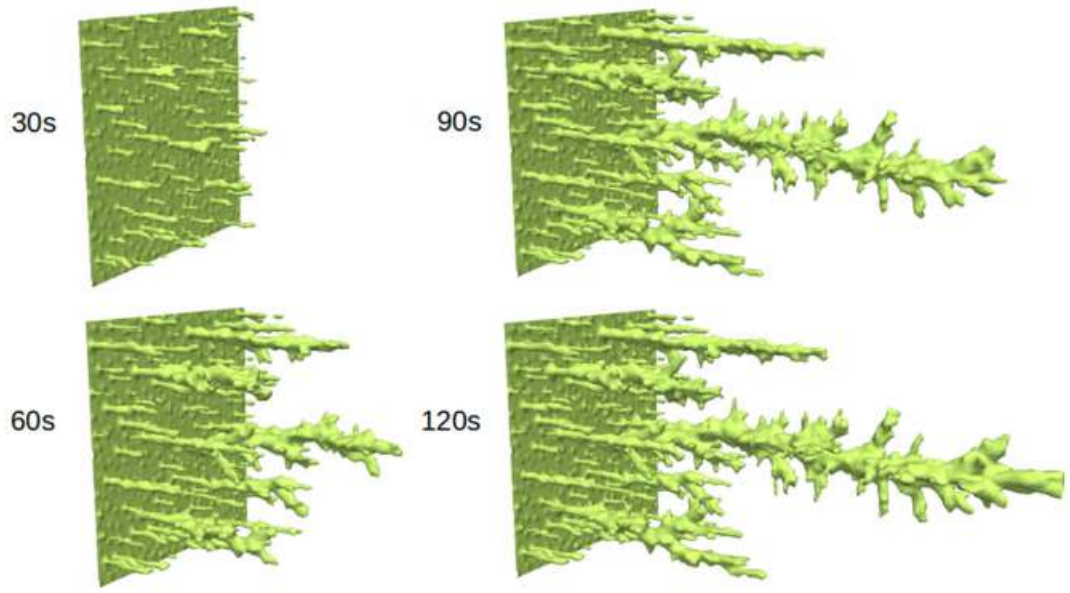


Figure 6.8: Simulation results of dissolution instabilities in a carbonate core. For convenience of the visualization, the cells that contain solid mineral are not displayed (from Soulaine and Tchelepi (2016)).

pattern almost reaches a steady-state, the channels get wider and smoother by the slow process of diffusion of the reactant in the matrix. This result is representative of the wormholes pattern obtained in Daccord and Lenormand (1987)'s experiments.

Our model can be used in subsurface engineering to determine the optimal injection rate to stimulate a clogged well or to increase the conductivity in deep geothermal reservoirs.

Microbially-induced calcite precipitation

Among the scientific literature on micro-continuum models for hydro-geochemical processes, the work of Minto and co-workers is of particular interest (Minto et al. 2019, 2018). They report leading-edge simulations of microbially-induced calcite precipitation. Microbially induced calcium carbonate precipitation (MICP) is a promising technique that could be used for soil stabilization, for permeability control in porous and fractured media, for sealing leaky hydrocarbon wells, and for immobilizing contaminants (Phillips et al. 2016).

They investigate optimum MICP treatment strategies by developing a micro-continuum model that includes bacteria transport and attachment, urea hydrolysis, tractable $CaCO_3$ precipitation, and modification to the porous media in terms of porosity and permeability. The transport of bacteria is modelled by an advection-dispersion-reaction equation that describes the evolution of the bacteria concentration including their attachment/detachment to the mineral surfaces. As other micro-continuum models, their approach describes processes both at the pore-scale using image-based simulations (see Fig. 6.9) and at the field-scale. Their model – conceptually validated against experimental data – narrows down the range of possible injection strategies to determine the most promising for implementation at a field scale. Their results indicate that phased injection strategies may lead to the

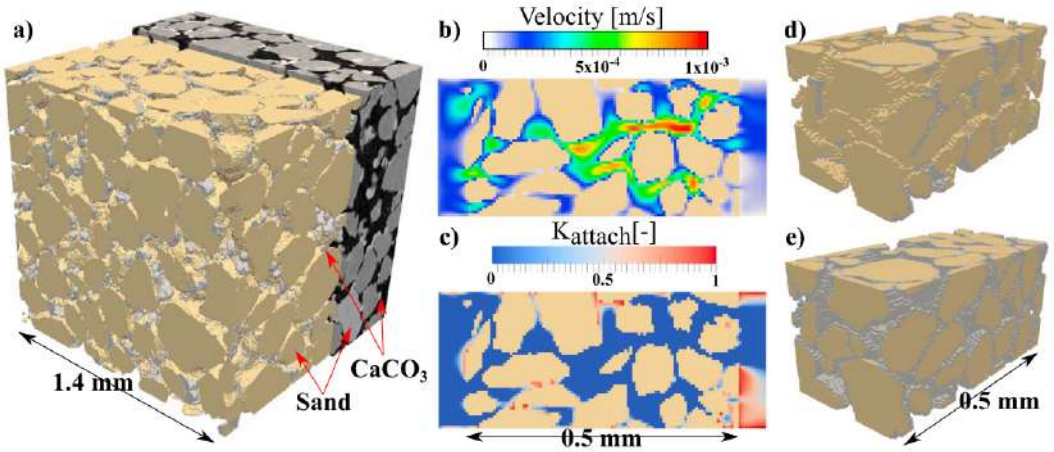


Figure 6.9: Microbially-induced calcite precipitation from Minto et al. (2019). (a) Greyscale X-ray CT data from a 6-mm-diameter core subsampled from a larger 40-cm-diameter block of MICP-treated beach sand. The data have been segmented based on X-ray attenuation to distinguish sand grains from $CaCO_3$. (b) Modelled velocity in a section (flow is from left to right) and (c) resulting velocity-dependent bacterial attachment coefficient. (d) $CaCO_3$ precipitation in model with velocity-dependent bacterial attachment, and, for comparison, (e) a model without.

most uniform precipitation in a porous medium.

Importantly, the authors emphasize the importance of an accurate description of bacterial attachment processes – at least the attachment rate should be a function of fluid velocity. Today, the modelling of attachment/detachment of all kind of colloidal particles (nanometals, fine particles, bacteria, viruses, asphaltenes..) in the subsurface remains a challenge that we are investigating in the PhD of Laurez Maya.

Modelling of biofilm growth

Micro-continuum models have an interesting potential for modelling biofilms growth and the subsequent pore-clogging in pore-scale micro-structures. In such cases, the biofilm is seen as a deformable solid phase that grows and moves with respect to the supply of nutriments (Tian and Wang 2019). The development of micro-continuum models for biofilms growth is an area of research that I would like to investigate in a near future.

6.5 Conclusion

Micro-continuum models are a versatile and powerful approach to simulate multi-scale coupled processes in porous media. The governing equations are rooted in the elementary physical principles and combined with appropriate sub-grid models for describing processes in the unresolved porosity. A key interest of the method is its ability to model pore-scale and field-scale processes within the same framework. State-of-the-art micro-continuum models handle two-phase flow, hydro-bio-geochemical processes, and poromechanics.

Despite its early age – the seminal work of Steefel et al. (2015) and Soulaine and

Tchelepi (2016) are barely half a decade old – micro-continuum approach for pore-scale processes has already demonstrated its strength in image-based simulations and coupled physics. The growing interest around micro-continuum approach is emphasized by the increasing number of scientific publications using such a concept.

On-going work is dedicated to the development of the resolved-unresolved interfacial conditions (e.g. for solute transport and multiphase flow), the modelling of colloidal particles attachment-detachment at the solid walls, and to the improvement of computational efficiency.

The next Chapter presents *porousMedia4Foam* our OpenFOAM® package based on micro-continuum concepts.

Chapter 7

porousMedia4Foam: the ultimate OpenFOAM package for solving flow and transport in porous media

porousMedia4Foam is the digital companion of this dissertation. porousMedia4Foam is our in-house package for solving flow and transport in porous media using the popular open-source numerical toolbox OpenFOAM®. The package includes the multi-scale micro-continuum models introduced in the former Chapter as well as more standard field-scale solvers. The code is in continuous development and is dedicated to coupled processes. Some snippets traced back to my PhD thesis (e.g. the two-phase flow solver impesFoam). The open-source aspect of porousMedia4Foam helps us to promote our new findings and to gather a community of users.

7.1 Introduction

porousMedia4Foam is a generic platform for solving flow and transport in porous media at various scales of interest. It is an open-source platform developed by the authors using the C++ library OpenFOAM (Chen et al. 2014a; Weller et al. 1998) that can be downloaded from http://www.openfoam.org. Hence, the package benefits from all the features of OpenFOAM including the solution of partial differential equations using the finite-volume method on three-dimensional unstructured grids as well as High-Performance Computing. *porousMedia4Foam* has multi-scale capabilities for solving two-phase flow (liquid-liquid and liquid-gas) in porous systems and hydro-geochemical processes.

The main architecture of the code has been developed by the author. Some of the codes and tutorials have been adapted from the *porousMultiphaseFoam* toolbox (Horgue et al. 2015) developed and maintained by my former colleague Pierre Horgue at Institut de Mécanique des Fluides de Toulouse. Others from the work of Francisco Carrillo at Princeton University and its *hybridPorousInterFoam* and *hybridBiotPimpleFoam* solvers[1] (Carrillo and Bourg 2019; Carrillo et al. 2020). Additional contributors include Saideep Pavuluri, Julien Maes, and Christophe Tournassat.

[1] https://github.com/Franjcf

7.2 Code architecture

The code is organized in three interacting parts: a class that describes porous media properties (Section 7.2.3), the flow solvers (Section 7.2.1) and the geochemical packages (Section 7.2.2). As *porousMedia4Foam* intends to be a versatile platform, it is designed in a such way that other porous media models, other flow solvers or other geochemical packages can be easily implemented using the C++ code architecture. In this section, we briefly introduce the models currently implemented in the code. More details are found in the related literature and in the previous chapters.

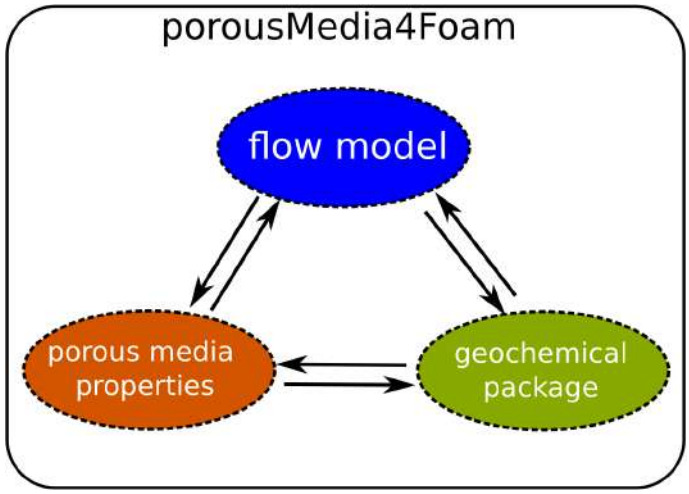


Figure 7.1: The code is organized in three interacting classes: a) porous media properties, b) the flow solvers, and c) the geochemical packages.

7.2.1 Flow solvers

porousMedia4Foam includes various flow models: multi-scale flow solvers based on the micro-continuum approach, continuum-scale Darcy solvers and a constant velocity solver (see Table 7.1). Each flow solvers is compiled into an executable that is run in a terminal to start a simulation. Each flow solver can call all the porous media models and geochemical libraries implemented in the package.

Table 7.1: Summary of the flow solvers implemented in *porousMedia4Foam*.

Name	Model	Comments
dbsFoam	Micro-continuum (Darcy-Brinkman-Stokes)	pore-scale, hybrid-scale, continuum-scale, Soulaine and Tchelepi (2016).
darcyFoam	Darcy's law	continuum-scale only.
impesFoam	Two-phase Darcy	continuum-scale only, Horgue et al. (2015)
hybridPorousInterFoam	Two-phase micro-continuum (Darcy-Brinkman-Stokes)	pore-scale, hybrid-scale, continuum-scale, Soulaine et al. (2019, 2018), Carrillo et al. (2020).
constantVelocityFoam	constant velocity profile	uniform or non-uniform velocity profiles.

7.2.2 Geochemical packages

In *porousMedia4Foam*, complex reaction networks are handled by geochemical packages (Soulaine et al. 2021b). The aqueous components are transported using the velocity profile, $\boldsymbol{v}_f$, computed by the flow solver (see section 7.2.1) and surface reactions rely on the reactive surface area, A_e, calculated with the porous media models (see section 7.2.3). The code architecture of *porousMedia4Foam* is generic so that a wide variety of third-party geochemical packages can be coupled with our platform for solving hydro-geochemical processes at the pore-scale and at the continuum-scale. For now, the popular geochemistry package PHREEQC (Parkhurst and Appelo 2013; Parkhurst and Wissmeier 2015) is used for complex hydro-geochemical simulations. Models currently implemented in *porousMedia4Foam* to account for geochemistry are summarized in Table 7.2.

Table 7.2: Summary of the geochemical packages implemented in *porousMedia4Foam* for simulating hydro-geochemical processes.

Name	Model	Comments
phreeqcRM	PHREEQC	Parkhurst and Wissmeier (2015)
simpleFirstOrderKineticMole	first order kinetic, C_i in mol/m^3	Molins et al. (2020), Soulaine et al. (2017b)
transportOnly	no geochemistry	–
flowOnly	no transport, no geochemistry	–

7.2.3 Porous media models

The flow solvers and geochemistry modules rely on porous media properties including absolute permeability, specific surface area and dispersion tensor. These properties describe pore-scale effects related to the micro-structure geometry of the porous medium. Hence, they may change if the micro-structure evolves with geochemical reactions.

Absolute permeability The absolute permeability describes the ability of a porous medium to conduct the flow. This property is intrinsic to the medium micro-structure and therefore evolves with geochemical processes including precipitation and dissolution. *porousMedia4Foam* includes several porosity-permeability relationships (e.g. Kozeny-Carman, power-law).

Dispersion tensor In porous media, the spreading of a solute is not governed only by molecular diffusion (D_i) but also by the micro-structure and the local velocity field. On the one hand, the tortuosity of the porous structure tends to slow down the spreading. On the other hand, hydrodynamic dispersion stretches a solute band in the flow direction during its transport. In *porousMedia4Foam*, a single effective diffusion tensor, D_i^*, is used to represent both mechanisms. Different dispersion tensor models are implemented in the code.

Surface area The estimation of the reactive surface area is crucial to model geochemical processes described by kinetic reactions. Actually, reactive surface area is a difficult quantity to assess as only a portion of the geometric surface area is accessible to the reactants. For example, in an advection-dominated transport, only the surfaces at the vicinity of the faster flowlines react (Soulaine et al. 2017b) leading to a reactive surface area smaller than the geometric surface area. Moreover, the evolution of the specific surface area as the mineral volume fractions change due to dissolution or precipitation is not necessarily monotonic (Noiriel et al. 2009). Various models are implemented in *porousMedia4Foam* to describe the surface area as a function of the mineral volume fraction.

Unlike continuum-scale simulations, the surface area in pore-scale modelling is not an input parameter but is a direct output of the simulation. Indeed at the pore-scale, the micro-structure of the porous medium is fully resolved in the computational grid and there is a sharp interface between the fluid and the solid mineral. The Volume-of-Solid model computes the surface area of an explicit fluid/solid interface using the gradient of the volume fraction of mineral (see Soulaine et al. (2017b) for additional details on the technique).

Relative permeabilities Two-phase flow simulations using either Darcy- or micro-continuum-based rely on the concept of relative permeabilities. The package includes the classic models by Brooks and Corey (1964) and van Genuchten (1980).

Capillary pressure curves Two-phase flow simulations using either Darcy- or micro-continuum-based rely on the concept of capillary pressure. The package includes the classic models by Brooks and Corey (1964) and van Genuchten (1980).

7.3 Installation instructions

The toolbox *porousMedia4Foam* only needs a standard OpenFOAM installation from `www.openfoam.org`. It has been tested on OpenFOAM v7 only. Geochemical packages such as PHREEQC are provided as Third Party.

To install *porousMedia4Foam*, download the latest version on our GitHub platform: `https://github.com/csoulain/porousMedia4Foam`.

Then, source the OpenFOAM configuration file (example for ubuntu version):

```
source /opt/openfoamv7/etc/bashrc
```

In the "porousMedia4Foam" directory, run :

```
./Allwmake
```

to install the package.

Dynamic libraries are compiled in the standard OpenFOAM user directory :

```
$FOAM_USER_LIBBIN
```

The executable solvers (e.g. impesFoam, dbsFoam, darcyFoam) are placed in the standard OpenFOAM user directory

```
$FOAM_USER_APPBIN
```

Each tutorial directory contains "run" and "clean" files to test installation and validate the solver.

To remove compilation and temporary files, run :

```
./Allwclean
```

More information about the toolbox is found in the ReleaseNotes.txt file and in the associated scientific publications (Carrillo et al. 2020; Horgue et al. 2015; Soulaine et al. 2021b) for detailed information about the toolbox.

Chapter 8

Conclusion and perspectives

Geological porous media are fascinating objects. In-depth knowledge of the transport processes is crucial to guide the exploitation of natural resources in the soils and the subsurface including energy resources (e.g. geothermal, oil and gas), the storage of gas (e.g. carbon dioxide, hydrogen) and nuclear waste disposals in deep geological formations, and the water resources management. Because of the time and spatial scales that characterize these exploitation projects, it is essential to develop predictive capacities to accompany the anthropogenic usage of the subsurface while minimizing its environmental impact. Indeed, the exploitation of deep resources involves drilling kilometer depth wells, and the nuclear waste disposals stored in argillaceous formations will stay there for thousands of years.

In a logic of cascade of scales nested within each other, *computational microfluidics for geosciences* appears as a crucial tool to investigate and decipher the coupled and complex processes occurring at the pore-scale in subsurface porous formations. Therefore, it is also an essential element to guide the derivation of robust and relevant field-scale models that are used in reservoir engineering. The concept of computational microfluidics developed by the author and its collaborators is the digital counterpart of microfluidic experiments. It augments laboratory experiments by providing dynamic mapping of velocity, pressure, aqueous concentration, and temperature profiles as well as the evolution of the solid mineral distribution, data that are not easily measurable in the lab. Computational microfluidics is also powerful to perform sensitivity analysis for identifying the key parameters governing the coupled processes. It also allows the exploration of ranges of pressure and temperature that cannot be achieved in the lab without dedicated equipment. Unlike pore-network modelling – a widely used approach for describing pore-scale physics – that relies on an approximation of the pore geometries and on pore-averaged physical laws, computational microfluidics solves the fundamental equations of continuum mechanics in the exact microstructure geometry. This means that computational microfludics is not only an ideal tool for upscaling purpose, but also a tool to investigate processes at the pore-scale.

State-of-the-art computational microfluidics for geosciences has the capabilities to model multiphase flow, mass transfer across moving interfaces (e.g. solubilization), and geochemical processes at the fluid-mineral boundary (e.g. dissolution, precipitation). Although this new discipline is still in its infancy, computational microfluidics has already brought strong scientific results. For example, it has been used successfully to assess the accessible reactive surface area according to flow con-

ditions. It has also highlighted the limiting role played by gaseous barriers in the dynamics of dissolution instabilities. Computational microfluidics is the physical engine of *Digital Rock Physics*, an emerging technology to compute rock properties (e.g. permeability, capillary pressure curves) by solving physical processes in three-dimensional image of rock sample. Simulation of larger and more diverse rock samples is a very attainable task due to the current continuous and massive growth of high-performance computing.

The capability of computational microfluidics to solve coupled processes is constantly improving. The micro-continuum approach introduced in this thesis allows computational microfluidics for solving coupled processes including multiphase flow, reactive transport, and poromechanics. This approach relies on the Darcy-Brinkman-Stokes equations, i.e. a locally averaged Navier-Stokes equations with appropriate subgrid models. Micro-continuum models are ideal to deal with transport in systems involving two characteristic pore sizes including fractured porous media for which the network of fractures is fully resolved while the porous matrix is described using homogenized concepts such as porosity, permeability, and specific surface area. Micro-continuum approaches for pore-scale modelling were initially designed in image-based simulations to compute the absolute permeability while accounting for the microstructure that was below the imaging instrument resolution, and, therefore, not observable in the image. Since this pioneering work, the approach keeps growing and the current models handle multi-phase flow in the presence of microporosity, displacement of fluid-mineral interfaces along with geochemical reactions (e.g. precipitation and dissolution), and the mechanical deformation of the solid matrix as in the swelling of clays. Far from being confined to small-scale systems, micro-continuum approaches are intrinsically multi-scale with the tremendous advantage that they are also valid for simulating coupled processes at the field scale. Hence, a unique system of partial differential equations is used for simulating coupled processes in porous media with characteristic sizes ranging from nanometers to kilometers and beyond.

The modelling approaches introduced in this thesis have been implemented in *porousMedia4Foam*, an open-source OpenFOAM® package for flow and transport in porous media developed by the author and its collaborators. The usage of this library goes way beyond geosciences as the models implemented are generic and apply to all kinds of fields involving porous materials being natural or manufactured (e.g. gas-liquid contactors in chemical engineering, effervescent tablet in drug delivery, biological tissues, coffee brewing, and batteries to name a few). The development of the modelling platform is always ongoing and includes the past, present, and future contributions of Ph.D. students, postdocs, and partners involved in my research.

In my future lines of research, I would like to understand how colloidal particles (e.g. nanoirons, bacteria) control two-phase flow in porous media. Deciphering these complex mechanisms is crucial to develop new remediation techniques for groundwaters contaminated with chlorinated solvents or hydrocarbon. This kind of pollution is recognized as among the most difficult to remove because droplets of contaminants are trapped in the pore space by capillary forces. The scientific strategy introduced in this thesis will allow an in-depth knowledge of the pore-scale processes leading to the remobilization of trapped droplets. Appropriate computational microfluidics will be a key asset to guide new remediation strategies. Their development necessitates pursuing the efforts that I am conducting for almost a decade.

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