AAP 2023 iMAT - Scientific application form

Seeing in the dark: Dynamics in opaque porous materials

Part 2: Project

2 a) Scientific context of project (max. 2,5 pages)

The goal of this Ph.D. project is to unravel the mechanisms of particle dynamics at micrometric scales in soils with X-ray imaging and modeling. We will first develop a new analysis tool to probe dynamical quantities from X-ray correlation microscopy in opaque heterogeneous materials in situ. We will then use the tool to image and understand particle dynamics in these materials, including active particle dynamics, in the global context of microorganism colonization of degraded soils.

Soils are mesmerizing materials in complexity, harboring a breadth of porous lengthscales, a diversity of compounds from organic to inorganic, and various forms of life. Already from a mechanical point of view, the arrangement of soil aggregates forms highly heterogeneous structures (Fig. 1a). Especially soil erosion by rainfall damage can cause soil crusting, forming a highly anisotropic, centimetric layer at the surface, which hinders the installation of new life forms [Pointing2012, Geoffroy2022]. Anisotropy creates labyrinthine networks inside the soil matrix, adding to the fascinating material properties [Dabat2019,Goral2022]. Such degraded soils are a crucial issue for sustainable agriculture and carbon sequestration in soils [Pointing2012,Jansson2020]. To ***foster the healing and re-colonization of degraded soils, it is key to understand how minuscule life forms, such as microorganisms, might navigate these environments*** and nest sustainably [Jansson2020].

The navigation pathway of microorganisms is microscopic [Raynaud2014,Pot2022]. However, the opaque nature of soils prohibits the use of traditional microscopic tools. Most studies thus focus on artificial porous media of packed transparent spheres [Bhattacharjee2019] or 2D geometries [Dentz2022,Kurz2022,Goral2022]. Such studies have unraveled that microorganism motion in porous media vastly differs from colloidal motion: dead-ends in the porous network act as traps [Bhattacharjee2019], and nematic order forces microorganisms to follow lines [Goral2022]. Yet, the inherent heterogeneity and anisotropy of natural soils, at multiple scales, can only affect this motion further. It is thus ***crucial to develop techniques to achieve in-situ measurements in opaque materials***.

Current approaches to understand navigation are often macroscopic, empiric, phenomenological, and cannot predict behavior over various soil compositions or conditions [König2020,Pot2022]. Since microscopic details are eluded, ***fundamental insight, i.e. the precise mechanisms by which transport is modified is still lacking*** [König2020,Dabat2020,Pot2022]. This means there are numerous open questions, especially, how do 3D anisotropy (Fig. 1b) and hierarchical organization of pores, modify colloidal (aggregate) or active (microorganisms) transport? Can we predict which key component of the porous network determines most crucially transport? What is the role of hydrodynamic effects?

The purpose of this Ph.D. is, on the one hand, to overcome transparency limitations and propose new techniques of experimental imaging in opaque materials. On the other hand, to determine the fundamental transport mechanisms of passive and active particles in such heterogeneous environments. The Ph.D. will combine exquisite multiscale modeling and X-ray imaging in collaboration between the lab PHENIX (Sorbonne Université) and the IC2MP (Université de Poitiers).

The first part of the Ph.D. will develop a robust imaging tool to extract dynamical properties from X-ray correlation microscopy. X-rays are especially suited for such opaque materials [Geoffroy2022]. Correlation methods, such as X-ray Photon Correlation Spectroscopy (XPCS) or Differential Dynamic Microscopy (DDM), are used in multiple contexts at various scales to probe dynamic properties [Rose2020]. Yet most of these techniques rely on beam scattering (DDM) [Cerbino2008], which is not accessible with X-rays, or coherent beams (XPCS) [Zinn2018] which is expensive and not suited for these time and length scales. In addition, most analysis tools are limited to acquiring *effective* quantities, such as the long-time diffusion coefficient or the average drift in the sample. In the presence of heterogeneities, such simple descriptions break, and new mathematical background is required. Our approach will build on a direct analysis of time-resolved 2D projections of the 3D porous network under consideration. As a preliminary step, we have demonstrated that temporal correlations of X-ray images acquired at SOLEIL synchrotron, numerically analyzed in the Fourier space, quantify the sedimentation speed of colloids inside a porous matrix (Fig. 1c). In this Ph.D. project, we will build a mathematical framework to quantify X-ray image correlations. We will start with 2D projections of diffusing monodisperse particles and build analytics with stochastic density functional theory [Dean1996] confirmed with Brownian Dynamics simulations. We will then extend the framework to bi-disperse and polydisperse particles, including immobile beads representing the porous matrix. Finally, we will study active particles, starting with an Active Brownian particle model. ***The development of this new methodology*** is essential to probe the kinetic response of particles in situ in such non-transparent materials. ***It will, in itself, be a significant contribution to the field*.**

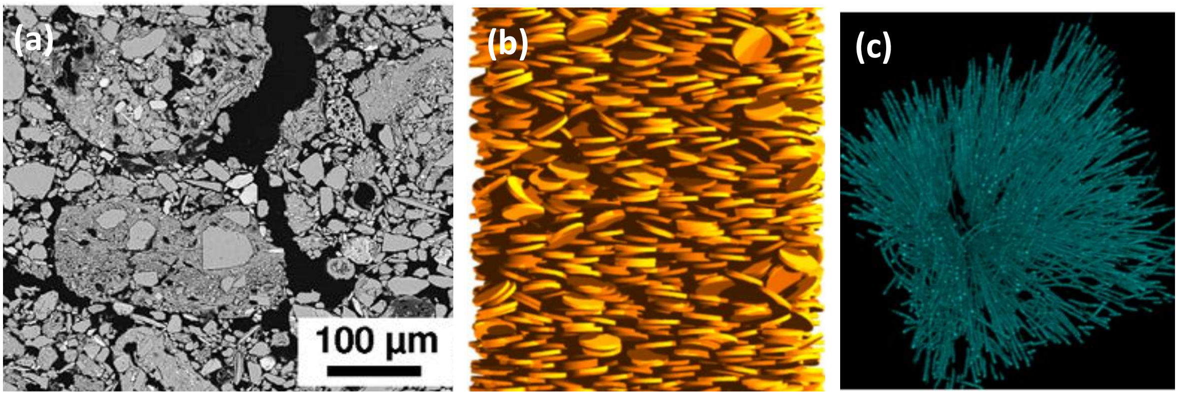

Figure 1: ***Structure and dynamics in highly heterogeneous materials*** (a) SEM image of topsoil aggregates [Geoffroy2022]; (b) Random close packing of anisotropic clay particles [Ferrage2015] (c) 3D X-ray reconstruction of colloidal sedimentation through the random close packing [Levitz 2019].

In the second part, we will harvest this new methodology to probe particle dynamics in increasingly complex artificial soils. We will start with a model porous matrix made of packed clay platelets (kaolinite), about 1-50µm, that we can design with different levels of anisotropy albeit with similar porosity [Dabat2020]. With X-rays, we will then investigate the motion of passive or active colloids, buoyant, about 1µm in size. Passive will be made of gold-coated (for more contrast) polystyrene spheres. Active Janus particles are hemispherically coated with platinum, with hydrogen peroxide in solution to propel, mimicking the motion of microorganisms [Becchinger2016]. This combination of sizes and materials guarantees that we will resolve the multiscale particle dynamics in space and time with current X-ray equipment in SOLEIL synchrotron. With this 1st set of experiments and the analysis tool developed in the first part, ***we will quantify how anisotropy and hydrodynamic interactions affect colloidal motion***, which is expected to be strong in the porous matrix [Dabat2019]. We will then design a 2nd set of experiments with more elaborate artificial soils, for example with thinner but more abundant particles, to amplify the effect of surfaces and labyrinthine layers. This is a key parameter since active particles tend to follow surfaces [Bhattacharjee2019]. We will thus be able to probe how much surfaces slow or accelerate colloids. To ***unravel the fundamental transport mechanisms***, ***we will taylor the Brownian Dynamics simulations*** of the first part, adding in tomography information of the actual porous matrix. Comparison with experiments will assess the impact of surfaces, hydrodynamic interactions, and activity on this heterogeneous anisotropy-modified transport.

Overall, we will construct **a global framework for in-situ observation of dynamics in opaque, highly heterogeneous model soil materials**. We will finally be able to extend it to ***natural soil samples in the final part,*** or as an important perspective of this Ph.D. Another essential aspect is that we will have assessed navigation laws of microorganisms in porous media, a key step for the re-colonization of degraded soils. From our navigation laws, we can extract likelihood distributions within the porous matrix, as a function of time. Adding reproduction rules or water localization in the matrix during droughts will pave the way to optimize colonization strategies [Raynaud2014].

Risks: Our in-situ approach with X-ray synchrotron imaging is ambitious and presents several challenging aspects such as (a) adjusting the experimental system for sufficient contrast and (b) analyzing the dynamics fast enough that we may resolve transport. ***We directly target these risks by developing a new analysis technique*** (1st part of the Ph.D.). Independently of its role in the project, this analysis tool will significantly contribute to the field, enabling the tracking of complex dynamics in opaque materials. In addition, ***we have designed bottom-up experiments***, starting with colloids in model soils – which have already been synthesized in our laboratory at a different – and going to active colloids in natural soils. If our approach reveals too challenging, we would track such particle dynamics in 2D micromodels, namely microfluidic arrays templated on natural soils [Roman2016].

Team: ***The supervising team is highly interdisciplinary and gathers all the required expertise for this ambitious project.*** The main advisor, Sophie Marbach, specializes in multiscale modeling [Cui2022]. She has recently developed an analytic tool to probe dynamic quantities from image correlations [Minh2023] and has expertise in porous media [Marbach2016, Marbach2018]. Fabien Hubert, the co-supervisor, has exquisite natural and artificial soil preparation ability [Bakker2019, Dabat2019]. Finally, Laurent Michot, co-supervisor, has long-standing expertise in X-ray imaging [Boucly2018, Ferrage2015]. Laurent and Fabien have recently jointly developed a new method to spatially resolve mineral concentrations with 2D X-rays [Geoffroy2022]. Additional collaborators include Pierre Levitz (PHENIX, Paris) for experimental analysis [Ferrage2015, Levitz2019]; Federico Paratore (ETH Zurich) for active colloids synthesis; Eric Ferrage (IC2MP, Poitiers) for simulations [Ferrage2015, Dabat2019].

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2 b) Research plan with provisional calendar (max. 0,5 page)

(year 1)
- Develop x-ray correlation microscopy
  - (i) Start with a monodisperse colloidal solution
  - (ii) Then bidisperse, polydisperse, with some immobile
  - (iii) Finally with Active Brownian particles
- *2 months stay in Poitiers* to synthesize initial model samples from kaolinite particles with 2 levels of anisotropy (isotropic or very anisotropic)
- In parallel, assemble material/synchrotron time grants for x-ray experiments
- Acquire expertise of active colloid synthesis (F. Paratore, ETH Zurich, collaborator of S. Marbach)

(year 2)
- Perform 1st set of Synchroton experiments with benchmark analysis + test upscale experiments
- *2 months stay in Poitiers* to synthesize 2nd set of model samples with varying surface ratio/anisotropy ratio.
- Perform 2nd set of Synchroton experiments with advanced samples

(year 3)
- assess current progress and risks. If necessary target 2D experiments or more modeling approaches.
- *1 month stay in Poitiers* to synthesize 3rd set of samples – if all goes well – take real soil samples
- (Final 6 months) Global conclusions of the work; prepare knowledge transfer
- (Final 4 months) PhD manuscript and defense.