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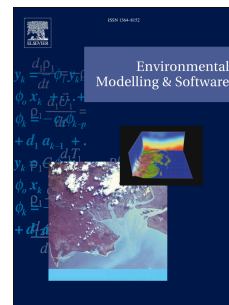


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1 *porousMedia4Foam*: Multi-scale open-source platform for 2 hydro-geochemical simulations with OpenFOAM[®]

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8 **Abstract**

porousMedia4Foam is a package for solving flow and transport in porous media using OpenFOAM[®] - a popular open-source numerical toolbox. We introduce and highlight the features of a new generation open-source hydro-geochemical module implemented within *porousMedia4Foam*, which relies on micro-continuum concept and which makes it possible to investigate hydro-geochemical processes occurring at multiple scales i.e. at the pore-scale, reservoir-scale and at the hybrid-scale. Geochemistry is handled by a third party package (e.g. PHREEQC) that is coupled to the flow and transport solver of OpenFOAM[®]. We conducted benchmarks across different scales to validate the accuracy of our simulator. We further looked at the evolution of mineral dissolution/ precipitation in a fractured porous system. Application fields of this new package include the investigation of hydro-bio-geochemical processes in the critical zone, the modelling of contaminant transport in aquifers, as well as and the assessment of confinement performance for geological barriers.

9 *Keywords:* Reactive Transport Modelling, Multi-scale simulations, Micro-continuum,
10 OpenFOAM, PHREEQC

11 **1. Introduction**

12 Over the last decades, reactive transport modelling (RTM) has become an essential tool
13 for the study of subsurface processes involving flow, transport and geochemical reactions
14 [1]. This discipline is at the junction of two scientific communities, namely geochemistry
15 and transport in porous media. RTM consists of computational models that describe the
16 coupled physical, chemical, and biological processes that interact with each other over a
17 broad range of spatial and temporal scales. RTM modelling tools enable the prediction of
18 contaminant migration in polluted aquifers, and are used to design enhanced remediation
19 techniques. Integration of physical and biogeochemical processes makes RTM also an ideal
20 research instrument for elucidating the complex and non-linear interactions between roots,
21 micro-organisms, water composition and minerals in the Critical Zone [2]. Other applications
22 include the assessment of the long-term integrity of reservoirs for storing carbon dioxide,
23 hydrogen or nuclear wastes in deep geological formations [3, 4]. In practice, three different

24 kinds of models are used to describe reactive transport in porous media as illustrated in
 25 Figure 1: (i) continuum models, (ii) pore-scale models, and (iii) hybrid models that combine
 26 both former approaches.

27 Continuum models (see Fig. 1c) are representative of the historical and standard ap-
 28 proach for solving reactive transport in large-scale natural porous systems [5]. Continuum-
 29 scale RTM codes include, among others, MIN3P [6], CrunchFlow [1], TOUGHREACT [7],
 30 PFlotran [8] or HP1 [9]. In such codes, flow and transport equations are formulated in terms
 31 of volume-averaged equations with respect to a Representative Elementary Volume (REV)
 32 of the porous structure [10] and are coupled with geochemical reactions (see Steefel et al. [1]
 33 for an comprehensive description of the coupling). The topology of the rock micro-structures
 34 is described using effective properties including porosity, tortuosity, permeability – or hy-
 35 draulic conductivity – and specific surface area. Flow is usually modelled using Darcy’s law
 36 and multi-component species transport relies on a set of advection-dispersion-reaction equa-
 37 tions. In addition to the classic challenges related to transport in porous media, including
 38 the medium heterogeneity awareness and the description of hydrodynamic dispersion, RTM
 39 has to consider the variation of rock properties in response to chemical reactions. Indeed,
 40 by enlarging or clogging pore throats and fractures, geochemical processes such as minerals
 41 dissolution and precipitation can alter the local flowfield and subsequently modify the rock
 42 properties, e.g permeability, tortuosity, accessible reactive surface area [11, 12].

43 Changes in rock properties with chemical reaction is usually described in continuum-
 44 scale RTM as heuristic functions of the porosity. For example, in most of reactive transport
 45 codes, tortuosity is described using Archie’s law, permeability change is modelled using
 46 Kozeny-Carman relationship, and mineral surface areas evolve as a two-third power law of
 47 the porosity [13]. However, the complex interplay between reactions, advection, and diffusion
 48 can lead to highly nonlinear porosity feedback that is poorly captured using this kind of
 49 relationships that were not built on a strong theoretical background [14, 15]. The limiting
 50 factor of the continuum-scale models is therefore the determination of empirical parameters
 51 and their evolution as a function of the progress of geochemical processes. To circumvent
 52 these challenges, recent efforts have focused on the numerical modelling of coupled hydro-
 53 geochemical processes at the pore-scale [16, 17, 18, 19, 20].

54 In pore-scale models (see Fig. 1a), the pore network is fully resolved, i.e. each point
 55 of space is occupied by either a fluid or solid phase. As the exact knowledge of the phase
 56 distribution is known, continuum-scale concepts such as porosity, permeability, and reactive
 57 surface area do not apply at the pore-scale. They can be obtained, however, by averaging
 58 pore-scale simulation results if the computational domain is large enough to reach the size
 59 of a REV [21, 22]. The strategy that consists in simulating flow and transport in a three-
 60 dimensional image of a porous sample to characterize its continuum-scale properties has
 61 become an independent scientific discipline sometimes referred to as Digital Rock Physics
 62 [23, 24, 25, 26]. Most of the efforts so far have been devoted to solve the Navier-Stokes
 63 equations under single [27, 28, 29, 30] and two-phase flow conditions [31, 32, 33, 34, 35]
 64 to compute absolute and relative permeabilities. Despite the growing investment in the
 65 development of RTM at the pore-scale – pioneer simulators date back to the late 90s [16] – the

66 pore-scale RTM field is still emerging. One of the main challenge of this approach consists in
67 moving the fluid/solid boundary with respect to chemical reactions at the mineral boundaries
68 [36]. A comprehensive review of the different approaches used to solve this problem can be
69 found in Molins et al. [37]. It is only very recently that pore-scale simulators have been proved
70 mature for reproducing accurately and without any adjusting parameters the dissolution of
71 a calcite crystal [38, 37], or of a gypsum grain [39]. Actually, the benchmark presented in
72 Molins et al. [37] is a first effort based on a relatively simple geochemical reaction (a single
73 component that reacts with a single mineral using a first-order kinetics) to demonstrate the
74 ability of current codes to accurately simulate mineral dissolution at the pore scale in a
75 reproducible manner with several codes. Further developments and verifications still need
76 to be done for simulating multi-component aqueous solutions interacting with heterogeneous
77 multi-mineral media using comprehensive reaction networks.

78 Naturally occurring porous media involve a wide range of spatial scales. For example,
79 the important contrast in pore-size distributions in fractured porous rocks comes from much
80 larger characteristic lengths for the fractures than for the surrounding porous matrix. There-
81 fore, the domain size required to reach a REV limits the use of pure pore-scale modelling.
82 Hybrid-scale models have been proposed to describe systems that include multiple charac-
83 teristic length-scales, for which some regions are described using pore-scale modelling while
84 others are modelled with continuum approaches (see Fig. 1b) [40, 41]. Two different ap-
85 proaches have been developed to solve hybrid-scale problems. On the one hand, the domain
86 decomposition technique solves different physics on separate domains – one for Darcy flow,
87 another for Stokes flow – linked together through appropriate boundary conditions [42] in-
88 cluding Beaver-Joseph and Ochoa-Tapia-Whitaker conditions [43, 44]. On the other hand,
89 micro-continuum models use a single set of partial differential equations throughout the com-
90 putational domain regardless of the content of a grid block [45, 46]. The latter approach is
91 particularly well-suited to capture the dynamic displacement of the interface between the
92 porous and solid-free regions without involving complex re-meshing strategies. For example,
93 micro-continuum models have been used successfully to simulate the formation and growth
94 of wormholes in acidic environments [47, 48, 46]. Hybrid-scale modelling is also a powerful
95 tool in image-based simulations to account for microscale features that are not visible in the
96 images because they are smaller than the imaging instrument resolution [49, 50, 51, 30, 52].

97 In this study, we developed a comprehensive open-source simulator to model coupled
98 hydro-geochemical processes at continuum-, pore- and hybrid-scales. This unique multi-
99 scale framework relies on the micro-continuum model and its ability to tend asymptotically
100 toward continuum-scale models if grid block contains solid content ($0 < \phi < 1$) or towards
101 pore-scale models otherwise ($\phi = 1$) [46]. The resulting advanced RTM allows the treat-
102 ment of complex reactions network as a function of flow conditions, water composition and
103 minerals distribution within the rock including the complex porosity feedback between flow
104 and chemistry. It is part of *porousMedia4Foam*, an open-source package developed by the
105 authors to solve flow and transport in porous media within the popular simulation platform
106 OpenFOAM®. Because of its versatility and advanced features such as three-dimensional
107 unstructured grids, dynamic meshes, and high-performance computing, there is a growing

108 interest in the community to develop mathematical models for solving flow and transport
 109 in porous media within OpenFOAM[®] [53, 54, 55, 38]. We developed a generic interface to
 110 combine flow and transport models with existing geochemical packages. We illustrate the
 111 versatility of our coupling interface by combining flow models with geochemical models us-
 112 ing PHREEQC [56], an open-source and popular geochemistry package that is used in many
 113 continuum-scale RTM [57, 58, 59, 60, 61].

114 In Section 2, we present the mathematical models implemented in *porousMedia4Foam*
 115 including a multi-scale and a continuum-scale flow solver, a wide range of porous properties
 116 models and the packages used to perform geochemical calculations. In Section 3, we verify the
 117 robustness of the coupled hydro-geochemical platform by simulating cases for which reference
 118 solutions exist both at the continuum-scale and at the pore-scale. Simulation results are
 119 compared with the results obtained with state-of-the-art reactive transport codes. Then in
 120 Section 4, we use the simulation framework to illustrate the potential of *porousMedia4Foam*
 121 to model hybrid-scale cases.

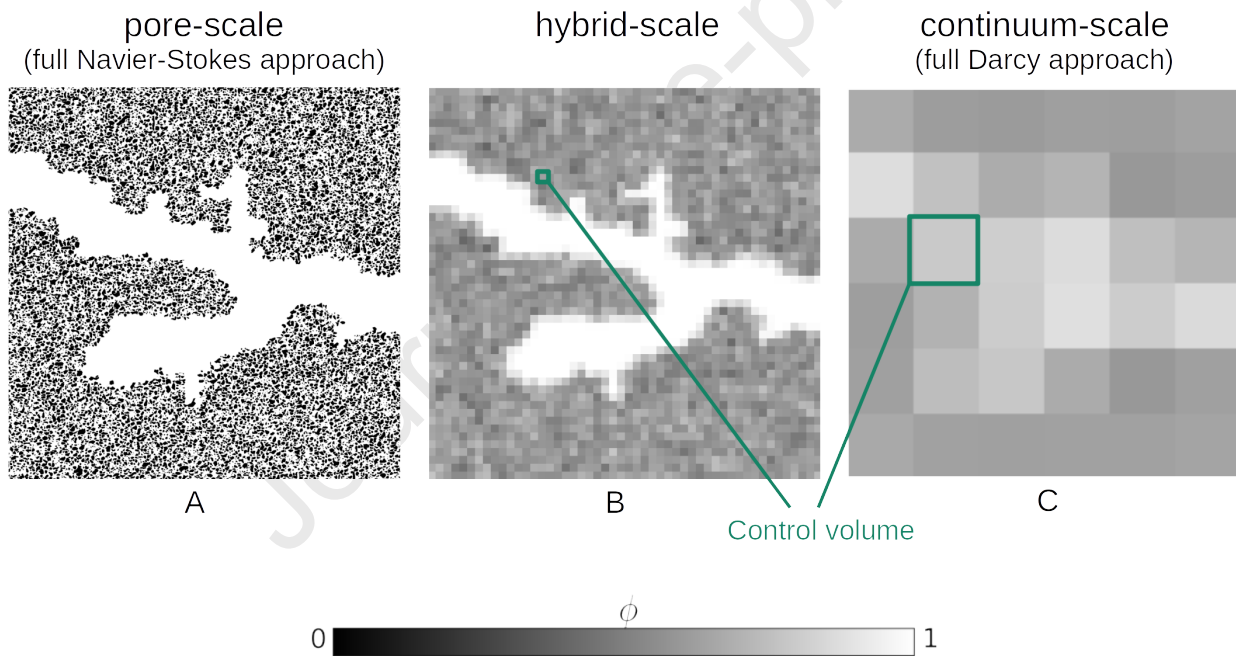


Figure 1: Porosity distribution considered in : *a*) a pure pore-scale approach (Navier-Stokes) for which the porosity is fully resolved, *b*) a micro-continuum approach (DBS) that handles region free of solid and porous region in the same framework, *c*) a pure continuum-scale approach (Darcy) for which all the control volumes contain an aggregate of fluid and solid.

122 2. The *porousMedia4Foam* package

123 The multi-scale solver for simulating hydro-geochemical problems is part of *porousMe-*
 124 *dia4Foam* (<https://github.com/csoulain/porousMedia4Foam>¹), a generic platform for
 125 solving flow and transport in porous media at various scales of interest. *porousMedia4Foam*
 126 is an open-source platform developed by the authors using the C++ library OpenFOAM
 127 (<http://www.openfoam.org>). Hence, the package benefits from all the features of Open-
 128 FOAM including the solution of partial differential equations using the finite-volume method
 129 on three-dimensional unstructured grids as well as High Performance Computing. Although
 130 *porousMedia4Foam* has capabilities for solving two-phase flow (liquid-liquid and liquid-gas)
 131 in porous systems, the geochemistry coupling that is introduced in this paper only considers
 132 single-phase flow.

133 The code is organized in three interacting parts: a class that describes porous media
 134 properties (Section 2.4), the flow solvers (Section 2.2) and the geochemical packages (Section
 135 2.3). As *porousMedia4Foam* intends to be a versatile platform, it is designed in a such way
 136 that other porous media models, other flow solvers or other geochemical packages can be
 137 easily implemented using the C++ code architecture. In this section, we introduce the
 138 models and their numerical implementation in the code. **It is organized as a multiple-entry**
 139 **document in which each model is presented in a self-contained manner so that users can**
 140 **directly refer to the subsection associated with the said model.**

141 2.1. Mineral distribution and porosity

A geological medium is made of an assembly of N_s minerals whose porous properties
 are defined in Section 2.4. The distribution of each mineral i on the computational grid is
 determined by the volume fraction,

$$Y_{s,i}(x, y, z, t) \text{ with } i \in [1, N_s], \quad (1)$$

142 in each grid block. The N_s solid volume fraction fields are dimensionless. They can be
 143 initialized with uniform or distributed values. The evolution of $Y_{s,i}$ due to geochemical
 144 reactions is dictated by the geochemical packages that are described in Section 2.3. In some
 145 simulations, it is relevant to define an inert mineral $Y_{s,\text{inert}}$ that is not part of the geochemical
 146 calculation.

The porosity field is computed by,

$$\phi = 1 - \sum_i^{N_s-1} Y_{s,i} - Y_{s,\text{inert}}, \quad (2)$$

147 and can be updated at any moment following dissolution or precipitation processes. **The**
 148 **porosity update at every time-step is optional.**

¹The link is deactivated at the moment. It will be activated as soon as the paper is accepted for publication.

149 *2.2. Flow solvers*

150 *porousMedia4Foam* for hydro-geochemical simulations includes three flow models: a
 151 multi-scale flow solver based on the micro-continuum approach, a continuum-scale Darcy
 152 solver and a constant velocity solver (see Table 1).

Table 1: Summary of the flow solvers implemented in *porousMedia4Foam* for simulating hydro-geochemical processes.

Name	Model	Section	Comments
dbsFoam	Micro-continuum (Darcy-Brinkman-Stokes)	Sec. 2.2.1	pore-scale, hybrid-scale, continuum-scale, Soulaire and Tchelepi [46].
darcyFoam	Darcy's law	Sec. 2.2.1	continuum-scale only.
constantVelocityFoam	constant velocity profile	Sec. 2.2.3	uniform or non-uniform velocity profiles.

153 *2.2.1. dbsFoam: Multi-scale micro-continuum flow model*

dbsFoam is a multi-scale flow solver based on the micro-continuum modelling approach developed in Soulaire and Tchelepi [46]. Micro-continuum approaches are intermediate between a pure Navier-Stokes description of the transport for which all the porosity is fully resolved (see Fig. 1a), and a pure continuum-scale modelling for which the flow is governed by Darcy's law (see Fig. 1c). This hybrid-scale approach relies on the Darcy-Brinkman-Stokes (DBS) equation [62] that allows for the modelling of flow and transport in regions free of solid and porous regions in a single framework [63, 46]. DBS equation arises from the integration of Navier-Stokes equations over a control volume [64, 65, 66, 67]. The momentum equation reads,

$$\frac{1}{\phi} \left(\frac{\partial \rho_f \mathbf{v}_f}{\partial t} + \nabla \cdot \left(\frac{\rho_f}{\phi} \mathbf{v}_f \mathbf{v}_f \right) \right) = -\nabla p_f + \rho_f \mathbf{g} + \nabla \cdot \left(\frac{\mu_f}{\phi} (\nabla \mathbf{v}_f + {}^t \nabla \mathbf{v}_f) \right) - \mu_f k^{-1} \mathbf{v}_f, \quad (3)$$

154 where ϕ is the porosity, \mathbf{v}_f is the seepage velocity, p_f is the fluid pressure, \mathbf{g} is the gravity,
 155 ρ_f is the fluid density, μ_f is the fluid viscosity and k is the cell permeability. The porous
 156 media properties including porosity and permeability change dynamically with geochemical
 157 processes and are updated at every time steps.

158 Eq. (3) is valid throughout the computational domain regardless the content of a cell.
 159 In regions that contain fluid only, $\phi = 1$, and the drag force $\mu_f k^{-1} \mathbf{v}_f$ vanishes so that
 160 the momentum equation tends to the Navier-Stokes equation. In regions that contain an
 161 aggregate of fluid and solid, $0 < \phi < 1$, and the drag force is dominant over the inertial and
 162 viscous forces so that Eq. (3) tends asymptotically to Darcy's law.

163 The momentum equation, Eq. (3), can be used to model pore-scale flows. Indeed, if
 164 a solid region is approximated by a low-permeability low-porosity matrix, the velocity in
 165 this region goes to near zero which forces a no-slip boundary condition at the fluid/solid
 166 interface. This feature of the DBS equation is particularly relevant to solve Navier-Stokes

167 problems using Cartesian grids only (also called penalized approach) [68, 46] and to move the
 168 fluid/solid interface with respect to geochemical processes such as precipitation/dissolution
 169 [38, 37] or swelling by using the local porosity field, ϕ , as a phase indicator function [69].

The pressure-velocity coupling is achieved by solving the momentum equation along with the micro-continuum continuity equation for multiple minerals. For an incompressible Newtonian aqueous fluid, the latter reads,

$$\nabla \cdot \mathbf{v}_f = \sum_{i=1}^{N_s} \dot{m}_{s,i} \left(\frac{1}{\rho_f} - \frac{1}{\rho_{s,i}} \right), \quad (4)$$

170 where $\rho_{s,i}$ is the solid density of mineral i and $\dot{m}_{s,i}$ represents the rate of phase change
 171 of solid into fluid, or of a fluid into solid. For example, it can represent the rate of solid
 172 minerals that is dissolved into aqueous solution. Inversely, it can describe an amount of
 173 fluid that is removed of a control volume because it has precipitated. The right-hand side
 174 of Eq. (4) is provided by the geochemistry calculation (Section 2.3). Although this term
 175 is often neglected in continuum-scale models, it ensures the mass balance at the fluid/solid
 176 interface in pore-scale simulations [38], as well as in continuum scale simulations [70].

177 The flow model formed by Eqs (3)-(4) is discretized using the finite volume method
 178 and solved sequentially. The pressure-velocity coupling is handled by a predictor-corrector
 179 strategy based on the PIMPLE algorithm implemented in OpenFOAM. It consists in a com-
 180 bination of PISO (Pressure Implicit with Splitting of Operator, Issa [71]) and SIMPLE (Semi-
 181 Implicit Method for Pressure Linked Equations, Patankar [72]). PIMPLE algorithm allows
 182 both transient and steady-state simulations. Moreover, PIMPLE enables larger time steps
 183 than PISO. Further information regarding the numerics is found in Soulaire and Tchepeli
 184 [46].

185 2.2.2. *darcyFoam*: Darcy's law

darcyFoam is a standard continuum-scale solver that is based on the Darcy's law,

$$\mathbf{v}_f = -\frac{k}{\mu_f} (\nabla p_f - \rho_f \mathbf{g}), \quad (5)$$

186 for describing flow in porous media. Numerically, Eq. (5) is combined along with Eq. (4)
 187 to form a Laplace equation solving implicitly for the pressure field, p_f . Then, the velocity
 188 field is calculated point-wise using Eq. (5) and p_f . If `activatePorosityFeedback` is switched
 189 on, Darcy's law is recalculated at every time steps to update the velocity and pressure fields
 190 according to the new permeability value.

Boundary conditions can be described by imposing fixed pressure or fixed velocity values on the domain edges. However, as *darcyFoam* solves implicitly for the pressure field, the boundary conditions on the velocity are transformed into pressure gradient conditions using Darcy's law:

$$\mathbf{n} \cdot \nabla p_f = -\mathbf{n} \cdot (\mu_f k^{-1} \mathbf{v}_f - \rho_f \mathbf{g}), \quad (6)$$

191 where \mathbf{n} is the vector normal to the domain boundary. In the code, Eq. (6) is achieved using
 192 the boundary condition `darcyGradPressure` [53].

193 2.2.3. *constantVelocityFoam*: Constant flow rate

194 The flow solver `constantVelocityFoam` is used to model cases in which the chemical species
 195 are transported using a steady-state velocity field, \mathbf{v}_f –uniform or non-uniform– provided
 196 as an input data that can come from a separate flow simulation. `constantVelocityFoam` is
 197 particularly useful if the feedback between geochemical reactions and the flow is negligible.
 198 Indeed, in such a case, the characteristic timescale of flow changes is much longer than the
 199 characteristic time of species transport and the calculation of the velocity profile can be
 200 **decoupled** from the species transport.

201 2.3. *Geochemical packages*

202 In *porousMedia4Foam*, complex reaction networks are handled by geochemical packages.
 203 The **aqueous components** are transported using the velocity profile, \mathbf{v}_f , computed by the
 204 flow solver (see section 2.2) and surface reactions rely on the reactive surface area, A_e , cal-
 205 culated with the porous media models (see section 2.4.2). The code architecture of *porous-*
 206 *Media4Foam* is generic so that a wide variety of third-party geochemical packages can be
 207 coupled with our platform for solving hydro-geochemical processes at the pore-scale and at
 208 the continuum-scale. In this paper, we illustrate the potential of the coupled simulation
 209 framework using the popular geochemistry package PHREEQC [73, 56]. Models currently
 210 implemented in *porousMedia4Foam* to account for geochemistry are summarized in Table 2.

Table 2: Summary of the geochemical packages implemented in *porousMedia4Foam* for simulating hydro-geochemical processes.

Name	Model	Section	Comments
phreeqcRM	PHREEQC	Sec. 2.3.1	Parkhurst and Wissmeier [56]
simpleFirstOrderKineticMole	first order kinetic, C_i in mol/m ³	Sec. 2.3.2	Molins et al. [37], Soulaine et al. [38]
transportOnly	no geochemistry	Sec. 2.3.3	–
flowOnly	no transport, no geochemistry	Sec. 2.3.4	–

The geochemical packages update the water composition, C_j and the distribution of the solid minerals $Y_{s,i}$, and return the rate of solid changes,

$$\dot{m}_{s,i} = -\frac{\partial \rho_{s,i} Y_{s,i}}{\partial t}, \quad (7)$$

211 where $\rho_{s,i}$ is the density of solid mineral i .

212 2.3.1. *phreeqcRM*

213 The `phreeqcRM` class calls the general-purpose geochemical reaction model PHREEQC
 214 through the `PhreeqcRM` module. It carries out the transport of the aqueous solution compo-
 215 sition, C_j (in mol/kg_{water}), along with equilibrium and kinetic reactions with the solid miner-
 216 als described by $Y_{s,i}$. `phreeqcRM` is set with components, i.e. `SOLUTION_MASTER_SPECIES`
 217 **total concentration**.

218 The geochemistry setup is carried out using an input file that follows PHREEQC format.
 219 Hence, the aqueous composition is defined in the block SOLUTION (0 for the composition
 220 of the injected fluid at the inlet boundary, 1 for the initial aqueous composition in the bulk).
 221 The EQUILIBRIUM_PHASES and KINETICS blocks are generated automatically within
 222 the code and the user only has to assign before the calculation which mode of reactions
 223 is used for each mineral. Moreover, *porousMedia4Foam* can load any customized database
 224 using PHREEQC format.

The coupling between transport and reactions relies on an operator-splitting approach based on the Strang's algorithm [74]. First, all species concentration fields, C_j , are transported sequentially using the advection-dispersion equations,

$$\frac{\partial \phi C_j}{\partial t} + \nabla \cdot (\mathbf{v}_f C_j) - \nabla \cdot (\phi \mathbf{D}_j^* \cdot \nabla C_j) = 0, \quad (8)$$

225 where \mathbf{v}_f is the fluid velocity computed with the flow solver (see Section 2.2) and \mathbf{D}_j^* is an
 226 effective diffusion tensor that accounts for tortuosity and hydrodynamic dispersion effects
 227 (see Section 2.4.3). The transport equation is discretized on the computational domain using
 228 the finite-volume method and solved implicitly using OpenFOAM's engines.

229 Then, the volume fractions of solid minerals, $Y_{s,i}$, are updated according to phase equi-
 230 librium and/or kinetic reaction calculations provided by PHREEQC. Reaction kinetics use
 231 the surface area computed at every time steps using the surface area models in section 2.4.2.
 232 It corresponds to the surface area per volume and its units are m^2/m^3 (or m^{-1}). Hence, the
 233 RATE block provided in PHREEQC database to describe reaction rates has to be defined
 234 accordingly.

235 2.3.2. *simpleFirstOrderKineticMole*

236 *simpleFirstOrderKineticMole* is a simple geochemical engine for solving the transport of a
 237 single species labelled A that reacts with solid minerals using first order kinetic reactions.
 238 It is an extension to multiple minerals of the model used in the benchmark presented in
 239 [37] in which pore-scale simulators were used to model the dissolution of a calcite crystal by
 240 hydrochloric acid.

The chemical reaction reads,



The mass balance equation for species A reads,

$$\frac{\partial \phi C_A}{\partial t} + \nabla \cdot (\mathbf{v}_f C_A) - \nabla \cdot (\phi \mathbf{D}_j^* \cdot \nabla C_A) = - \left(\sum_{j=1}^{N_s} A_{s,j} (k_{j,A} \gamma_A) \right) C_A, \quad (10)$$

241 where \mathbf{v}_f is the fluid velocity, \mathbf{D}_j^* is an effective diffusion tensor, $A_{s,j}$ is the reactive surface
 242 area of mineral j (in m^{-1}), and $(k_{j,A} \gamma_A)$ is the constant of reaction of the species A with
 243 the mineral j (in m/s) following the notations adopted in [37]. In *simpleFirstOrderKinetic-*
 244 *Mole*, the concentration field, C_j , is defined in mol/m^3 . The equation is discretized on the
 245 computational grid using the finite-volume method and solved implicitly.

The distribution of solid minerals evolves according to,

$$\frac{\partial Y_{s,i}}{\partial t} = -A_{s,A} (k_{i,A} \gamma_A) V_{m,s,i} C_A, \quad (11)$$

246 where $V_{m,s,i}$ (in m^3/mol) is the molar volume of the reacting mineral.

247 2.3.3. *transportOnly*

transportOnly solves the advection-dispersion equation,

$$\frac{\partial \phi C_j}{\partial t} + \nabla \cdot (\mathbf{v}_f C_j) - \nabla \cdot (\phi \mathbf{D}_j^* \cdot \nabla C_j) = 0, \quad (12)$$

248 without considering geochemistry. It allows the transport of species using the dispersion
249 models implemented in *porousMedia4Foam* (see Table 5).

250 2.3.4. *flowOnly*

251 *flowOnly* is an empty class for computing velocity profiles without species transport nor
252 geochemistry. For example, Poonosamy et al. [11] used the multi-scale flow solver of *porous-*
253 *Media4Foam* to compute the steady-state velocity profile in absence of geochemical reactions
254 within a two-scale domain, i.e. a domain that contains both solid-free regions and porous
255 regions (see Fig. 1b). This option is particularly interesting in cases for which geochemistry
256 and flow can be treated independently from each other.

257 2.4. Porous media models

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

262 2.4.1. Absolute permeability models

263 The absolute permeability describes the ability of a porous medium to **conduct** the flow.
264 This property is intrinsic to the medium micro-structure and therefore evolves with geochem-
265 ical processes including precipitation and dissolution. *porousMedia4Foam* includes several
266 porosity-permeability relationships summarized in Table 3.

267 2.4.2. Surface area models

268 The estimation of the reactive surface area is crucial to model geochemical processes
269 described by kinetic reactions. Actually, reactive surface area is a difficult quantity to assess
270 as only a portion of the geometric surface area is accessible to the reactants. For example,
271 in an advection-dominated transport, only the surfaces at the vicinity of the faster flowlines
272 react [38] leading to a reactive surface area smaller than the geometric surface area. Moreover,
273 the evolution of the specific surface area as the mineral volume fractions change due to
274 dissolution or precipitation is not necessarily monotonic [79]. Table 4 summarizes the models

Name	Expression	Comments
none	$k = 0$	–
constant	$k = k_0$	k_0 is uniform or non-uniform.
Power-law	$k = k_0 \left(\frac{\phi}{\phi_0}\right)^n$	n is a user defined variable.
Kozeny-Carman	$k = k_0 \left(\frac{\phi}{\phi_0}\right)^n \left(\frac{1-\phi_0}{1-\phi}\right)^m$	by default, $n = 2$ and $m = 3$ [75, 76].
Verma-Pruess	$k = k_0 \left(\frac{\phi-\phi_c}{\phi_0-\phi_c}\right)^n$	n is a model parameter. ϕ_c refers to the critical porosity where permeability reduces to 0 [77].
Hele-Shaw	$k = \frac{h^2}{12}$	for simulating 2D depth-averaged flow in micromodels (e.g. Poonosamy et al. [11], Roman et al. [78]).

Table 3: Summary of the permeability-porosity models implemented in *porousMedia4Foam*. In the table, subscripts 0 refers to variable data at initial time. Optionally, ϕ_0 and k_0 are updated at every time-steps.

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 Soulaire et al. [38] for additional details on the technique).

2.4.3. Dispersion models

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. The models implemented in the code are summarized in Table 5.

3. Verification of the hydro-geochemical simulation platform

In this section, the multi-scale hydro-geochemical simulation package *porousMedia4Foam* introduced in Section 2 is used along with PHREEQC to investigate several scenarios for which reference solutions exist. The verification of the results is achieved by comparison against benchmarks published in literature both at the continuum-scale and at the pore-scale. Essential files required to run all the test cases presented in this section are available as examples within the package. All simulations were run on Intel Xeon with 2.60 GHz.

Table 4: Summary of the specific surface area models implemented in *porousMedia4Foam*. Units of specific surface area are m^{-1} .

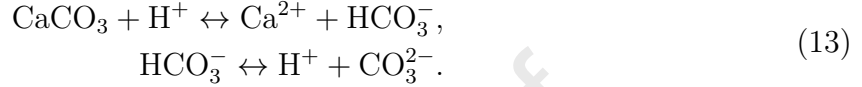
Name	Expression	Comments
None	$A_s = 0$	If specific surface area is not necessary, e.g. for phase equilibrium calculation.
Constant	$A_s = A_0$	–
Volume of solid	$A_s = \nabla Y_s \psi$	for pore-scale simulations only. Compute the local surface area based on the mineral mapping. ψ is a diffuse interface function [38].
Power-law	$A_s = A_0 (Y_s)^n$	n is a user defined variable
Sugar-lump	$A_s = \left(A_0 + A_m \left(1 - \left(\frac{Y_s}{Y_0} \right)^{n_1} \right)^{n_2} \right) \left(\frac{Y_s}{Y_0} \right)^{n_3}$	Evolution of the surface area of an aggregate during dissolution [79]. A_m is the maximum surface area given by the sum of the surface areas of all individual particles, n_1, n_2 and n_3 are user-defined parameters.
Hydro-geochemical coupling	$A_s = A_0 \left(\frac{Y_s}{Y_0} \right)^n (1 - \exp(-Pe^{-p} Da^{-q}))$	including surface reduction due to hydro-geochemical coupling [38]. n, p, q are user defined parameters.

Table 5: Summary of the dispersion models implemented in *porousMedia4Foam*. \mathbf{I} is the unit tensor.

Name	Expression	Comments
none	$D_i^* = 0$	for modelling transport by advection only
diffusionOnly	$D_i^* = D_i \mathbf{I}$	no tortuosity effects, no hydrodynamic dispersion.
archiesLaw	$D_i^* = \phi^n D_i \mathbf{I}$	tortuosity is represented by ϕ^n . By default, $n = 0$
linearDispersion	$D_i^* = \phi^n \left((D_i + \alpha_T \mathbf{v}) \mathbf{I} + \frac{(\alpha_L - \alpha_T)}{ \mathbf{v} } \mathbf{v} \mathbf{v} \right)$	tortuosity is represented by ϕ^n . α_L and α_T are model parameters describing lateral and longitudinal dispersion respectively.

297 *3.1. Verification at the continuum-scale*

We verify the ability of *porousMedia4Foam* to simulate coupled hydro-geochemical processes that include porosity feedback on the transport properties at the continuum-scale. We also verify that the multi-scale solver *dbfFoam* tends asymptotically towards Darcy’s law in porous domains. The case is based on the Benchmark 1 described in Xie et al. [13]. It consists of a 2 meters long 1D column initially filled with 35 % of inert mineral and 30 % of calcite. An acid ($\text{pH} = 3$) is continuously injected at the inlet to initiate the dissolution of calcite according to,



298 Table 6 provides the initial and boundary conditions data specific to the primary components.
299 A difference of 0.007 m in hydraulic head is applied between the inlet and outlet [13] by fixing
300 the pressure at 70 Pa and 0 Pa respectively at the inlet and outlet boundaries throughout the simulation.

Primary components	Units	Initial conditions	Boundary condition
pH	-	9.38	3
-	-	9.38	3
Ca	mol/kg _{water}	1.56×10^{-4}	9.97×10^{-5}
C(4)	mol/kg _{water}	2.56×10^{-4}	9.97×10^{-3}
S(6)	mol/kg _{water}	9.97×10^{-11}	6.44×10^{-4}

Table 6: Initial and boundary conditions of the primary components for calcite dissolution under kinetic conditions considering porosity feedback.

301
302 The calcite dissolution with porosity feedback is simulated using both the multi-scale
303 solver *dbfFoam* and the continuum-scale solver *darcyFoam*. The aqueous species are trans-
304 ported by advection only. The calcite dissolution is modelled using a kinetic rate of reaction
305 with $k_{\text{calcite}} = 5 \times 10^{-5}$ mol/m²/s and the initial specific area $A_0 = 1$ m²/m³. As the calcite
306 dissolves, the surface area decreases according to a power-law function with $n = 2/3$ (see
307 Table 4). The porosity-permeability relationship is described by the Kozeny-Carman equa-
308 tion (Table 3). The initial permeability of the column is set to $k_0 = 1.186 \times 10^{-11}$ m². The
309 column is spatially discretized with $\Delta x = 25$ mm (80 cells). 150 years are simulated with
310 $\Delta t = 21600$ s.

311 There is a perfect match between *dbfFoam* and *darcyFoam* confirming that the multi-scale
312 solver converges well to the continuum-scale solution predicted by Darcy’s law (Figure 2).
313 The analysis of results includes the evolution of porosity (Fig. 2a), calcite volume frac-
314 tion (Fig. 2b), hydraulic head (Fig. 2c) along the column length and the outflux over time
315 (Fig. 2d). To be consistent with the benchmark of [13], we consider the cross-sectional area
316 of the column to be 1 m². As the dissolution of calcite occurs, the calcite volume fraction
317 decreases and the porosity increases over time. In Fig. 2c, we notice different slopes of hy-
318 draulic head at different times. The slope is minimal where porosity is large and vice-versa.

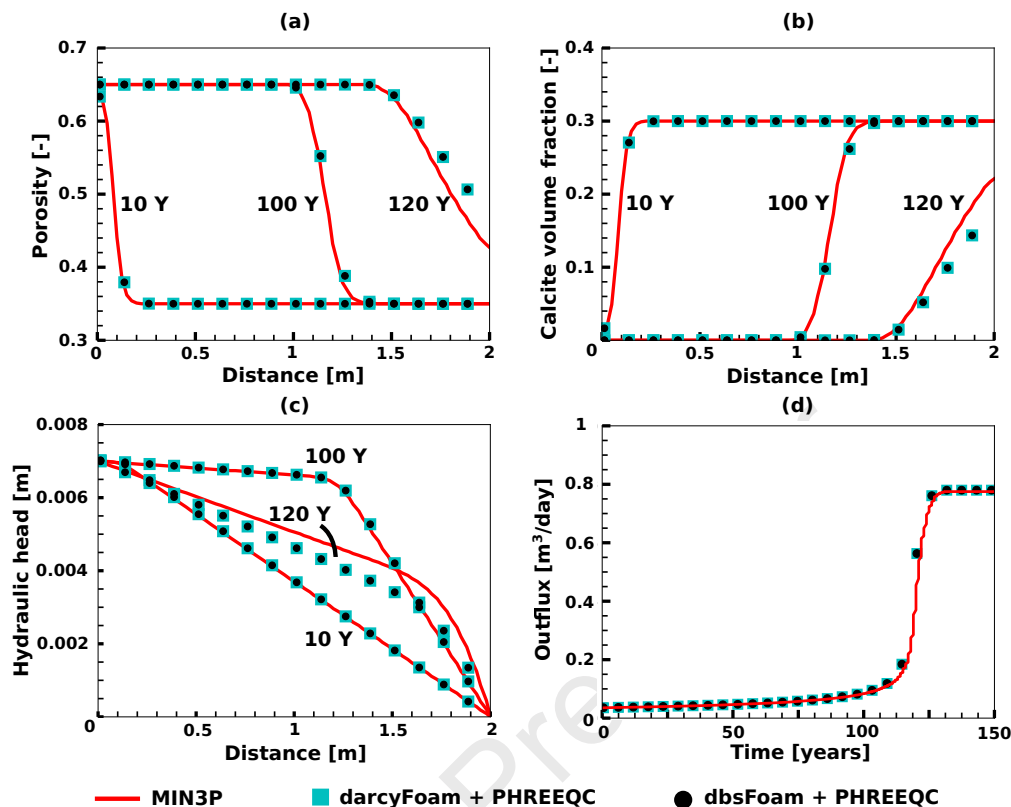


Figure 2: Calcite dissolution under kinetic conditions considering feedback of porous media properties. Evolution of (a) porosity, (b) calcite volume fraction, (c) hydraulic head along the channel and (d) evolution of outflux. MIN3P data is from [13] for comparison.

319 The velocity – and therefore the outflux – increases over time as the permeability (and poros-
 320 ity) of the system increases. The evolution of porosity, calcite volume fraction, hydraulic
 321 head and outflux predicted by *porousMedia4Foam* solvers are in close agreement with those
 322 of MIN3P which demonstrates the ability of our platform to simulate hydro-geochemical
 323 processes with porosity feedback.

324 3.2. Verification at the pore-scale

325 In this section, we highlight the capabilities of our OpenFOAM package to model hydro-
 326 geochemical interactions occurring at the pore-scale using PHREEQC.

In *porousMedia4Foam*, pore-scale simulations are run using the micro-continuum ap-
 proach through the flow solver *dbsFoam*. At the pore-scale, the reaction rates are directly
 applied at the fluid-mineral interface that is described explicitly in the computational grid
 using the mineral volume fraction. The micro-continuum approach has been used to sim-
 ulate the dissolution of a calcite crystal at the pore-scale and compared successfully with
 microfluidic experiments [38]. In Molins et al. [37], the approach is compared with state-of-
 the-art RTM at the pore-scale using various numerical techniques including Chombo-Cruch
 with Level-Set [20], Lattice Boltzmann Method [80], *dissolFoam* moving grids with confor-

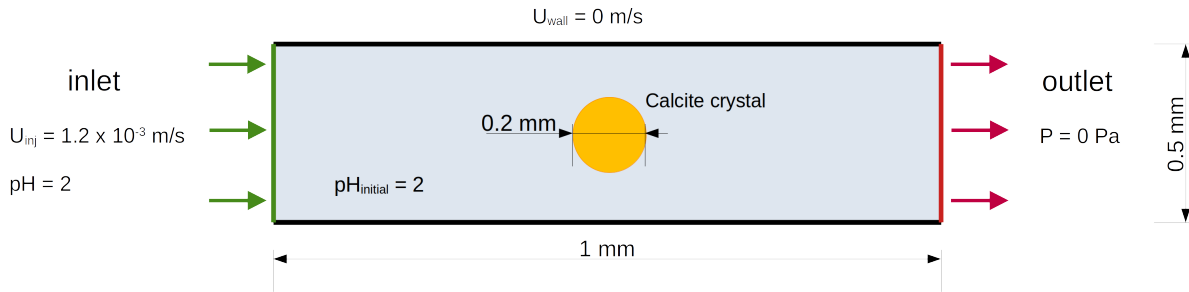


Figure 3: The considered model set-up along with initial and boundary conditions to investigate calcite grain dissolution in a microchannel.

mal mapping [81], and Vortex methods [82]. The benchmark consists of a 0.2 mm diameter calcite crystal posted in a 1 mm long 0.5 width channel (see Fig. 3). An acidic solution of pH=2 is continuously injected from the inlet at a rate of $U_{inj} = 1.2 \times 10^{-3}$ m/s. The calcite crystal dissolution is described considering a kinetic rate,

$$r = A_{\text{calcite}} (k_{\text{calcite}} \gamma) c_{H^+}, \quad (14)$$

where r is the reaction rate in mol/m³/s, A_{calcite} is the reactive surface area in m²/m³ computed using the volume-of-solid approach (see Table 4), $(k_{\text{calcite}} \gamma) = 0.89 \times 10^{-3}$ m/s is the reaction rate constant of calcite and, c_{H^+} is the concentration of H^+ in mol/m³. This reaction rate may not be fully representative of the underlying geochemical processes. It has been chosen in Molins et al. [37] to demonstrate the ability of various approaches to move the fluid-mineral interface according to geochemical processes. All the numerical methods were able to capture accurately the shape evolution of the calcite crystal, giving confidence in pore-scale simulators for moving fluid-mineral interfaces along with geochemical processes.

Actually, in Soullaine et al. [38] and Molins et al. [37], the micro-continuum approach, `dbsFoam`, was combined with the geochemical package `simpleFirstOrderKineticsMole` (see Table 2) that solves Eq. (14) using OpenFOAM's internal engines. This limits drastically the applicability of the approach to comprehensive reaction networks. In this section, we reproduce the two-dimensional case presented in Molins et al. [37] using `dbsFoam` and `phreeqcRM` to demonstrate the robustness of our coupling between OpenFOAM and PHREEQC at the pore-scale. The kinetic rate in PHREEQC input file has been modified to match Eq. (14). The system is spatially discretized using a Cartesian mesh of 128×64 cells. The simulation is run for 45 minutes using a time step size $\Delta t = 5$ ms.

The shape evolution of the calcite grain determined by the two approaches matches perfectly (Fig. 4) which verifies, therefore, that in our modelling platform, PHREEQC can be used to model hydro-geochemical interactions occurring at the pore-scale. This gives us confidence for further investigations that rely on more complex reactive transport phenomena occurring at the pore-scale.

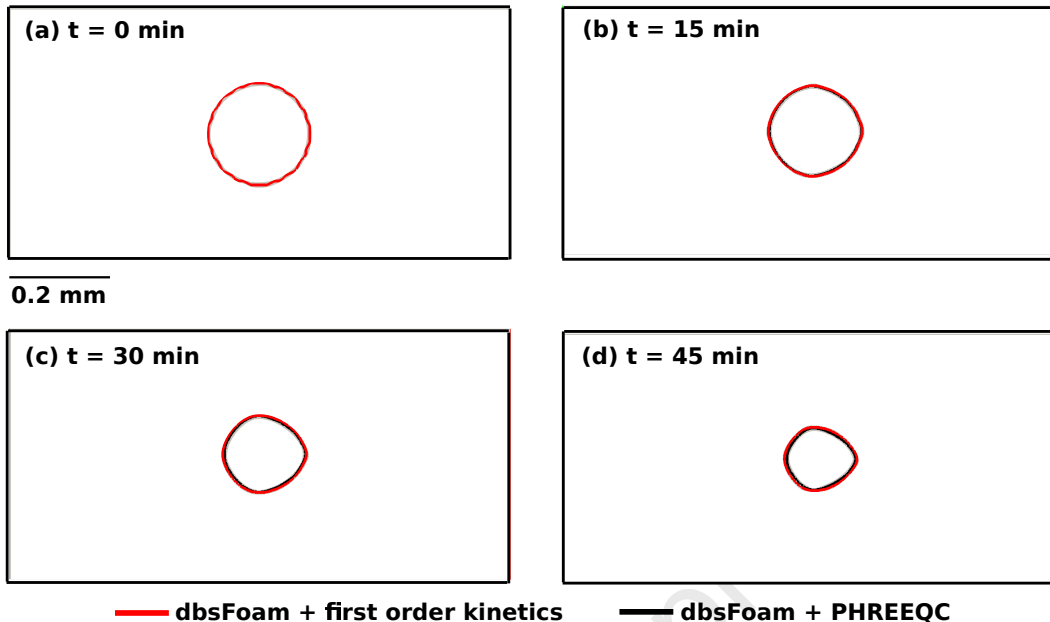


Figure 4: Evolution of the shape of the calcite grain as a function of time ((a) $t = 0$ min, (b) $t = 15$ min, (c) $t = 30$ min and (d) $t = 45$ min) predicted by `dbsFoam` + first order kinetics geochemical module (`simpleFirstOrderKineticsMole`, red line) and `dbsFoam` + PHREEQC (`phreeqcRM` geochemical module of `porousMedia4Foam`, black line). The red and black lines represent cells having calcite volume fraction of 0.5.

349 4. Hybrid-scale simulation in fractured porous media

350 In most subsurface environments, fractures intercept porous media domains. These fractures
 351 act as free-flow zones transporting substantial quantities of fluids alongside chemical
 352 species compared to the flow and transport that occur within the porous medium [83, 84].
 353 The complex interplay between advection, diffusion, and reaction can lead to very different
 354 dissolution and precipitation patterns. For example, Poonoosamy et al. [11] uses micro-
 355 Raman spectroscopy to visualise the replacement of celestite with barite in a fractured
 356 porous media flooded with a solution that contains barium ions. They observe that the
 357 mineral replacement occurs either uniformly or at the vicinity of the fracture-matrix inter-
 358 face. This difference in the mineral distribution was attributed to the injection flow rates
 359 leading to advection or diffusion-dominated transport.

We investigate such a multiscale system, where a fracture is sandwiched in between a reactive porous matrix made of 50% celestite (SrSO_4) having specific reactive surface area of $A_0 = 20000 \text{ m}^2/\text{m}^3$ as shown in Fig. 5. The fracture has a length of $\ell = 0.03 \text{ m}$ and height $h = 0.002 \text{ m}$. Transport phenomena in the fracture is fully resolved, i.e. the flow is governed by Navier-Stokes equations whereas the flow in the matrix is described by Darcy's law. This hybrid-scale case is modelled using the `dbsFoam` solver. The initial porosity and permeability of the porous medium are $\phi_0 = 0.5$ and $k_0 = 10^{-12} \text{ m}^2$, respectively. A solution containing $300 \text{ mol}/\text{m}^3$ of barium (Ba^{2+}) is continuously injected through the inlet at a constant velocity U_{inj} for 200 hours. The dispersivity of species within the porous matrix are

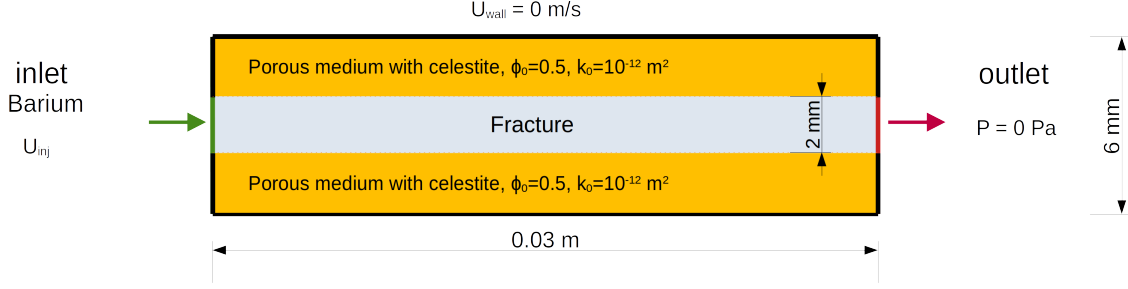
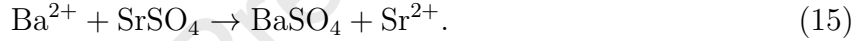


Figure 5: Numerical set-up for the hybrid-scale case study. A fracture is sandwiched in between two layers of reactive porous medium. The reactive porous medium comprises of celestite. Barium is injected at the inlet. We investigate this scenario considering two different injection velocities, $U_{inj} = 10^{-2} \text{ m/s}$ and $U_{inj} = 10^{-6} \text{ m/s}$.

taken into account considering linear dispersion law (Table 5) with molecular diffusion set to $D_j = 10^{-9} \text{ m}^2/\text{s}$, hydrodynamic dispersion coefficient set to $\alpha_L = 10^{-5} \text{ m}$ and tortuosity exponent set to $n = 2$. Once the barium ions reach the porous matrix, celestite dissociates into strontium (Sr^{2+}) and sulphate (SO_4^{2-}) ions. The barium ions react with sulphate ions resulting in the precipitation of barite (BaSO_4) according to the following reaction [85],



360 Celestite dissolution is taken into account considering kinetics with $k_{\text{celestite}} = 10^{-5.66} \text{ mol/m}^2/\text{s}$
 361 whereas, barite precipitation is accounted considering phase equilibrium. Celestite reactive
 362 surface area evolves linearly with its volume fraction. The matrix permeability is updated
 363 according to Kozeny-Carman.

364 We investigate the ongoing hydrogeochemistry within this system considering two dif-
 365 ferent injection velocities, $U_{inj} = 10^{-2} \text{ m/s}$ and $U_{inj} = 10^{-6} \text{ m/s}$. The Peclet number, $Pe =$
 366 $U_{inj}\ell/D_j$ (where the reference length-scale is the fracture aperture), characterizes the im-
 367 portance of advection with respect to diffusion within the fracture. The highest velocity
 368 corresponds to advection-dominated transport ($Pe \approx 10^4$) while the lowest corresponds to
 369 diffusion-dominated regime ($Pe \approx 1$). For both cases, the second Damkhler number that
 370 determines the timescale of reaction with respect to the timescale of species diffusion at the
 371 mineral surface is $Da_{II} = k_{\text{celestite}}\ell/(c_{\text{Ba}^{2+}}D_j) \approx 3.6 \times 10^{-4}$ (where the reference length-scale
 372 is the inverse of the specific surface area, $\ell = A_0^{-1}$, according to Soulaire et al. [38]).

373 We notice differences in the pattern of celestite dissolution and barite precipitation
 374 whether the transport in the fracture is dominated by advection or by diffusion in agreement
 375 with Poonosamy et al. [11] observations (Fig. 6). For advection dominated regime ($Pe > 1$),
 376 there are two characteristic timescales for the solute transport: first, the barium ions flow
 377 through the fracture by advection, then they diffuse laterally into the matrix. Because of
 378 the timescale contrast between the two processes, the concentration profile of barium ions
 379 is uniform along the porous matrix which leads to a uniform pattern of celestite dissolution
 380 and barite precipitation as seen in Figs. 6 and 7. For diffusion-dominated regimes ($Pe \leq 1$),
 381 the characteristic transport timescales both in the fracture and in the matrix are of the
 382 same order of magnitude. Therefore, the front of barium ions in the matrix follows the

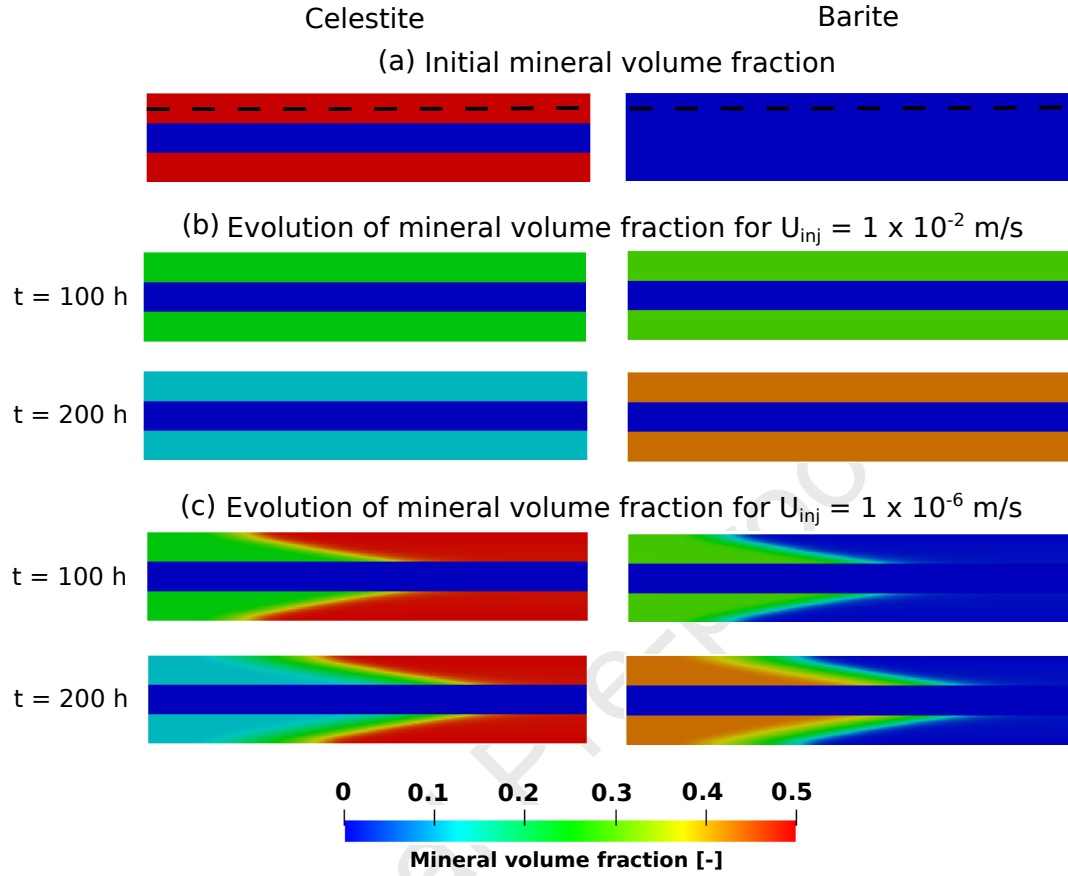


Figure 6: Evolution of mineral volume fractions - celestite on the left and barite on the right - at different injection velocities. (a) Initial ($t = 0$ s) mineral volume fractions. (b) Mineral volume fractions at 100 h and 200 h for $U_{inj} = 1 \times 10^{-2}$ m/s, and (c) mineral volume fractions at 100 h and 200 h for $U_{inj} = 1 \times 10^{-6}$ m/s.

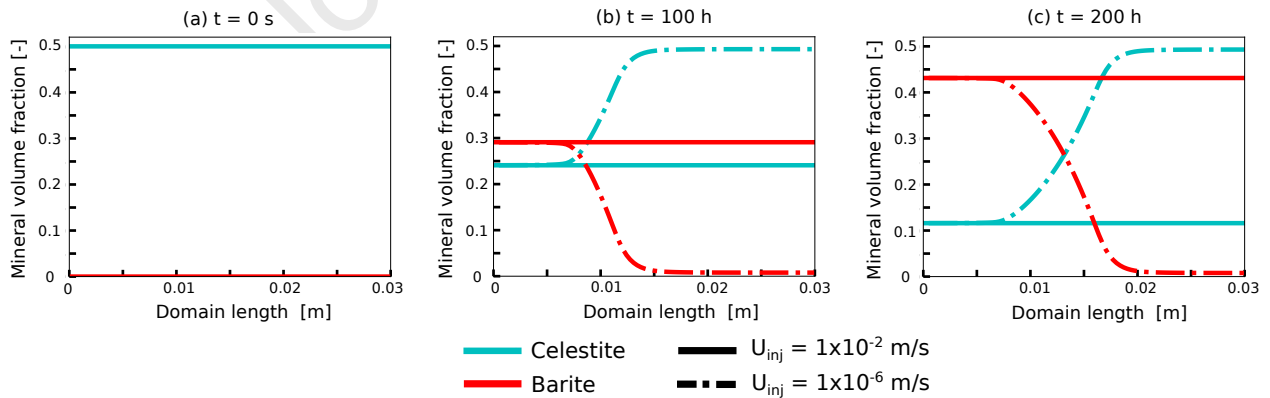


Figure 7: Plots comparing the mineral volume fractions of celestite and barite at three different time intervals (a) $t = 0$ s, (b) $t = 100$ h and (c) $t = 200$ h for different injection velocities. The mineral volume fraction data is collected along the length of the domain at a distance of 0.001 m from the top wall boundary as highlighted by dashed black line in Fig. 6 (see initial mineral volume fraction).

diffusive front within the fracture. Subsequently, we observe mineral dissolution (celestite) and precipitation (barite) fronts within the porous matrix (Figs. 6 and 7).

This illustration emphasizes the capabilities of *porousMedia4Foam* to model dual-porosity systems in reactive environments using hybrid-scale approach. Our platform is therefore a powerful tool to complement and augment reactive transport experiments including high-resolution imaging of the evolution the fracture aperture including the effect of the weathered zone [83, 79, 84, 86] and two-scale reactive microfluidic experiments [11, 87, 88].

In a logic of cascade of scales nested within each other, fractured porous media can be modelled by: *i)* pore-scale approaches, *ii)* discrete fracture networks, *iii)* dual porosity models. The hybrid-scale approach that we propose in this paper is intermediate between a full pore-scale description in which the fracture and the pores in the matrix are fully resolved and a discrete fracture network in which the matrix is modelled as a porous medium and the fractures as discrete elements that exchange matter with the matrix. The hybrid-scale approach is therefore crucial to characterize and improve the effective parameters (e.g. transfer functions between the porous matrix and the fracture) used in discrete fracture networks and larger-scale models.

5. Conclusion

We developed an integrated open-source simulator to model hydro-geochemical processes at various scales of interest including pore-scale and reservoir-scale. The simulation platform is part of *porousMedia4Foam*, a package that solves flow and transport in porous media using the open-source library OpenFOAM. The modelling framework handles complex reactions network as a function of flow conditions, water composition and minerals distribution within the rock including the complex porosity feedback between flow and chemistry. Moreover, *porousMedia4Foam* benefits from all features of OpenFOAM libraries. Hence, the code is fully parallel and handles structured as well as unstructured grids in one, two and three dimensions. The interface between the flow simulator and the geochemistry is generic and can be used to couple a large variety of geochemical packages. In this paper, we illustrated the hydro-geochemical capabilities of the coupled solver using PHREEQC.

Unlike other reactive transport simulators, *porousMedia4Foam* is multi-scale, i.e. a unique flow solver describes transport processes both at the continuum-scale and the pore-scale. Importantly, the two scales can be solved simultaneously in geological formations that feature large contrast of permeability and porosity. For example, in fractured rocks, *porousMedia4Foam* solves Stokes flow in the fracture network and Darcy's law in the porous matrix. This multi-scale model is achieved using the micro-continuum approach, hybrid-scale approach based on the Darcy-Brinkman-Stokes equation. Indeed, this approach is intermediate between a pure Navier-Stokes description of the transport for which all the porosity is fully resolved and pure continuum-scale modeling based on Darcys law. Besides this hybrid-scale approach, *porousMedia4Foam* also includes a standard Darcy solver for continuum-scale simulations. Therefore, the same simulator can be used to simulate flow, transport, and geochemical reactions in an reservoir and in 3D micro-tomography images.

423 The coupled hydro-geochemical simulator was verified by running cases for which refer-
424 ence solutions exist. These solutions are well-established and used in the reactive transport
425 community to benchmark state-of-the-art codes available both at the continuum-scale [13]
426 and at the pore-scale [37]. Finally, we demonstrated the ability of our advanced modelling
427 framework to simulate dissolution and precipitation processes in fractured porous media at
428 the pore-scale using the hybrid-scale approach. Here, the reactive medium consisted of ce-
429 lestite grains that reacted with a barium chloride solution injected into the system, leading
430 to the dissolution of celestite and the growth of barite. We observed differences in mineral
431 precipitation - dissolution patterns by varying the injection rates.

432 Because *porousMedia4Foam* has already capabilities for modelling two-phase flow in
433 porous media both at the pore- and Darcy's scales using two-phase micro-continuum tech-
434 nique [52, 89, 90], true multiscale and multiphase RTM is envisioned to be implemented in
435 *porousMedia4Foam* framework.

436 Software and Data Availability

437 *porousMedia4Foam*, the software introduced in this paper is built using open-source li-
438 braries including OpenFOAM and PHREEQC. The source code and the cases presented in
439 the paper are available on GitHub ([https://github.com/csoulain/porousMedia4Foam²](https://github.com/csoulain/porousMedia4Foam<sup>2</sup)).

440 Contributions of the authors

441 CS is the *porousMedia4Foam* architect. CS and SP implemented new models in *porous-*
442 *Media4Foam* and corrected bugs. SP and CT designed and setup the benchmark problems.
443 SP run the simulations. CS, SP, CT and FC discussed, interpreted the results and wrote the
444 paper. CS, CT and FC applied for funding.

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²The link is deactivated at the moment. It will be activated as soon as the paper is accepted for publi-
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Dear Editor,

We declare no conflict of interest.

Best regards,

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Highlights

- We developed an open-source platform for multi-scale simulations of hydro-geochemical processes,
- A unique formulation can handle pore-scale, hybrid-scale and continuum-scale simulations for hydro-geochemical processes,
- The framework is verified using reference solutions both at the pore-scale and at continuum-scale using PHREEQC,
- We investigate reactive transport in a fractured porous medium at the hybrid-scale.

Declaration of interests

■ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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