



# Reactive transport under stress: Permeability evolution in deformable porous media



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## ABSTRACT

We study reactive transport in a stressed porous media, where dissolution of the solid matrix causes two simultaneous, competing effects: pore enlargement due to chemical deformation, and pore compaction due to mechanical weakening. We use a novel, mechanistic pore-scale model to simulate flooding of a sample under fixed confining stress. Our simulations show that increasing the stress inhibits the permeability enhancement, increasing the injected volume required to reach a certain permeability, in agreement with recent experiments. We explain this behavior by stress concentration downstream, in the less dissolved (hence stiffer) outlet region. As this region is also less conductive, even its small compaction has a strong bottleneck effect that curbs the permeability.

Our results also elucidate that the impact of stress depends on the dissolution regime. Under wormholing conditions (slow injection, i.e. high Damkohler number,  $Da$ ), the development of a sharp dissolution front and high porosity contrast accentuates the bottleneck effect. This reduces transport heterogeneity, promoting wormhole competition. Once the outlet starts eroding, the extreme focusing of transport and hence dissolution—characteristic of wormholing—becomes dominant, diminishing the bottleneck effect and hence the impact of stress at breakthrough. In contrast, at high flow rates (low  $Da$ ), incomplete reaction upstream allows some of the reactant to traverse the sample, causing a more uniform dissolution. The continuous dissolution and its partial counteraction by compaction at the outlet provides a steady, gradual increase in the effect of stress. Consequently, the impact of stress is more pronounced at high  $Da$  during early stages (low permeability), and at low  $Da$  close breakthrough. Our work promotes understanding of the interplay between dissolution and compaction and its effect on the hydromechanical property evolution, with important implications for processes ranging from diagenesis and weathering of rocks, to well stimulation and carbon sequestration.

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## 1. Introduction

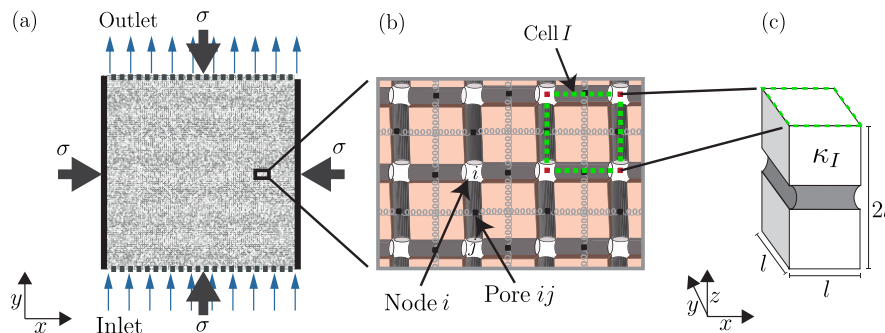
The transport of reactive fluids in porous media is a fundamental process in many earth systems, including weathering and diagenesis of rocks, contaminant transport, subsurface remediation, hydrocarbon recovery, and CO<sub>2</sub> sequestration. Reactive transport is a complex, nonlinear process: the transport and reactive properties and hence the rate and spatial distribution of fluid flow and reaction strongly depend on the microstructure, which in turn keeps evolving in time with the reaction (Dentz et al., 2011; Steefel and Maher, 2009; Noiriél, 2015).

Much interest has been given to the dissolution of the solid matrix and the consequent evolution of the porosity and permeability.

A rich range of behaviors (regimes) has been shown to emerge, depending on the interplay between advection, diffusion, and reaction. This interplay can be characterized by a pair of dimensionless numbers: (i) the Damkohler number,  $Da$ , describing the relative rate of reaction vs. advection; and (ii) Peclet,  $Pe$ , the ratio of advection vs. diffusion, or, equivalently, their product  $PeDa$  (reaction vs. diffusion, termed Peclet–Damkohler or kinetic number) (Bekri et al., 1995; Golfier et al., 2002; Steefel and Maher, 2009). It has been shown that for slow reactions (relative to the flow rate, small  $Da$ ), ample reactant is provided throughout the sample, leading to relatively spatially-uniform dissolution, and steady enhancement of permeability (Bekri et al., 1995; Golfier et al., 2002; Szymczak and Ladd, 2009). In contrast, at higher  $Da$  (the so-called transport-limited regime), most of the reactant is consumed by preferential dissolution of the most conductive flow paths (“wormholes”). This further enhances their conductivity relative to other, less conductive regions, leading eventually to runaway permeabil-

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**Fig. 1.** Schematics of our pore-scale model of reactive transport in a stressed, deformable porous medium. We capture the coupling between chemical and mechanical deformation through a discrete representation, combining: (i) pore network modeling of reactive transport, with (ii) interacting network of linear elastic blocks. (a) We simulate the injection of a reactive fluid into a sample held under fixed isotropic stress,  $\sigma$ . The sample is square, with two open faces (inlet and outlet, in the  $y$  direction) and two impermeable ones (in  $x$ ). (b) The pore space is discretized into a 2-D regular network of cylindrical pores connected by volumeless nodes (or junctions), squeezed in between solid blocks. Heterogeneity is provided by variations in pore sizes. (c) The basic model unit (“cell”) includes a solid block, with four engraved “half-channels”. The network properties are updated at each time step considering both the chemical and mechanical effects of dissolution.

ity increase (Daccord and Lenormand, 1987; Bekri et al., 1995; Hoefner and Fogler, 1988).

In many cases, the medium is under stress, such that dissolution (“chemical deformation”) also induces mechanical deformation. Chemomechanical deformation and the coupled changes in the transport and mechanical properties is key in many applications, such as enhanced hydrocarbon recovery and carbon geosequestration (Croizet et al., 2013; Rohmer et al., 2016; Carroll et al., 2016). Considerable attention has been given to the study of rough fractures, due to the importance of fracture flow and their relatively simple quasi two-dimensional (2-D) geometry (e.g. Liu et al., 2006; Detwiler, 2008; Elkhoury et al., 2013; Ishibashi et al., 2013; Ameli et al., 2014). Fewer works addressed porous media such as soils (Cha and Santamarina, 2016) and rocks (Zheng and Elsworth, 2012; Croizet et al., 2013; Emmanuel et al., 2015; Buscarnera and Das, 2016; Nguyen et al., 2016; Rohmer et al., 2016; Liu and Mostaghimi, 2017), mostly focusing on the evolution of the mechanical properties.

The evolution of transport properties has been recently studied experimentally, in rock cores subjected to external stress and flushed by a reactive fluid (see Vanorio, 2015; Clark and Vanorio, 2016 and the references therein). These experiments show considerable reduction in mechanical stiffness, and inhibition of permeability enhancement by compaction, in spite of net removal of solid mass. A model providing fundamental understanding of the interplay between dissolution and mechanical compaction and its effect on the permeability evolution, is lacking, providing the motivation for this paper. Existing models of stressed rock dissolution either address much larger scales (reservoir) (Nguyen et al., 2016), much smaller scales (describing changes to the local microstructure but not to permeability, a macroscopic property) (Emmanuel et al., 2015), or consider other mechanisms for porosity reduction such as pressure solution and re-precipitation (Zheng and Elsworth, 2012).

A major challenge towards understanding chemomechanical deformation is its dependency on pore-scale mechanisms and heterogeneity, inherent in geologic media. While the importance of pore-scale processes has long been recognized, rigorous studies are only recently made possible owing to advancements in experimental and computational capabilities, as highlighted in a recent special volume of review papers (Steeffel et al., 2015). The natural complexity of pore-scale processes associated with heterogeneity makes characterization (Noiriél, 2015; Steefel et al., 2015, and the references therein) as well as modeling and interpretation (Molins, 2015; Yoon et al., 2015; Xiong et al., 2016; Smith et al., 2017; Steefel et al., 2015) extremely challenging, in particular when mechanical deformation is involved (Emmanuel et al., 2015; Cha and

Santamarina, 2016; Liu and Mostaghimi, 2017). Despite of the advancements in experimental techniques, numerical simulations remain highly attractive (Molins, 2015; Yoon et al., 2015; Xiong et al., 2016; Liu and Mostaghimi, 2017; Smith et al., 2017).

In this paper, we present a novel pore-scale model, which, to the best of our knowledge, is the first to describe the permeability evolution caused by coupling between dissolution and compaction in a porous medium. Our simulations show that increasing stress inhibits the permeability evolution, requiring longer times or larger volumes injected to reach a certain permeability, in agreement with experiments. We also expose the interplay with the flow rate ( $Da$ ), that is the different effect of stress in wormholing vs. a more uniform dissolution regime. We find that at high  $Da$ , stress acts to reduce transport heterogeneity, promoting wormhole competition.

## 2. Pore-scale model

### 2.1. Conceptual model and computational approach

We simulate the injection of a reactive fluid into a porous medium held under a fixed confining stress. Our model accounts for the gradual, continuous dissolution of the solid matrix, and its coupling with mechanical weakening and compaction, excluding deformation by mechanisms such as fracturing (Røyne and Jamtveit, 2015; Clark and Vanorio, 2016), detachment of mineral grains (Mangane et al., 2013; Clark and Vanorio, 2016), and pressure solution and welding of microporous zones (Ishibashi et al., 2013; Emmanuel et al., 2015; Clark and Vanorio, 2016). Seeking fundamental understanding rather than quantitative prediction for a specific type of medium, we use here a simplified analog model medium: a 2-D network of cylindrical channels between soluble solid blocks. The coupling between dissolution and compaction at the pore-scale is captured by combining two discrete, overlapping representations: (i) pore network modeling of reactive transport and (ii) a network of interacting linear elastic porous solid blocks. Each discrete model unit (termed “cell” hereafter) includes one solid block, bounded by four “half-channels”—the respective void space associated with that cell (Fig. 1). Considering such model medium greatly simplifies the computation of fluid transport, reaction and chemomechanical deformation. This allows us to use sufficiently large domains required to capture the pattern evolution, repeated over multiple realizations (different samples with similar statistical properties) essential for systematic, mechanistic understanding in light of the sensitivity to the microstructural details. While it is possible to accurately represent the microstructure of natural geologic media with models such as direct numerical simulations or Lattice-Boltzmann (Yoon et al., 2015; Molins, 2015;

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