Microfluidic experiments of dissolution in a fracture. Influence of Damköhler and Péclet numbers, and of the geometry on the dissolution pattern

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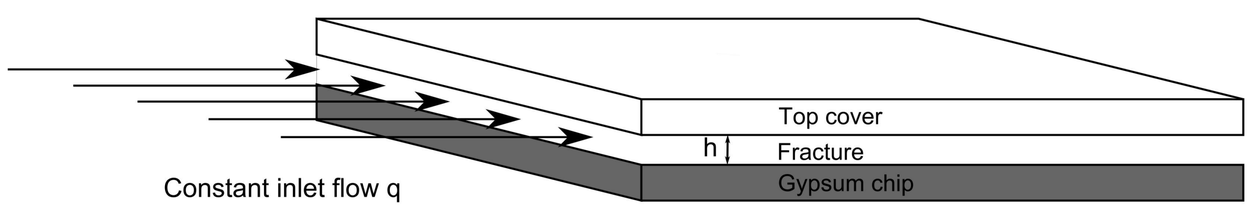
Introduction

Dissolution of natural rocks is an ever present phenomenon in nature. The shaping of natural landscapes by the dissolution of limestone gives for example birth to exceptional features like karsts. Currently, dissolution is also at the heart of key research topics such as Carbon Capture and Storage or Enhanced Oil Recovery. The basic principles of dissolution are well-known, however, the sheer amount of different patterns arising from these mechanisms and the strong dependency on parameters such as pore network, chemical composition and flow rate, make it particularly difficult to study theoretically and experimentally. In this study we present a microfluidic experiment simulating the behavior of a dissolving fluid in a fracture. The experiments consist of a chip of gypsum inserted between two polycarbonate plates and subjected to a constant flow rate of pure water ($CaSO_4,2H_2O = Ca^{2+} + SO_4^{2-} + 2\ H_2O$). The point in using microfluidics is that it allows a complete control on the experimental parameters such as geometry and chemical composition of the porous medium, flow rate, fracture aperture, roughness of the fracture walls, and an *in situ* observation of the geometry evolution which is impossible with 3D natural rocks. The purpose of the presented study is to experimentally map the patterns of dissolution as a function of the two fundamental parameters Damköhler and Péclet number and to compare it with numerical simulations.

Theoretical background and experimental set-up

Reactive flow in a fracture



Pressure	p
Solute concentration	c
Concentration at saturation	c_{sat}
Diffusion coefficient	D
Porosity	ϕ
Molar Volume	υ

Constitutive equations

Incompressible flow $\nabla \cdot q = 0$

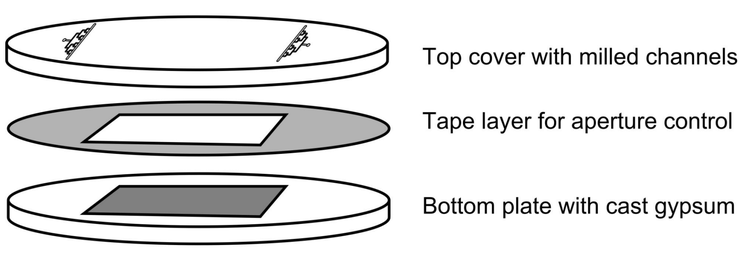
Flow in a Hele-Shaw cell $q = -\frac{h^3}{12\eta}\nabla p$

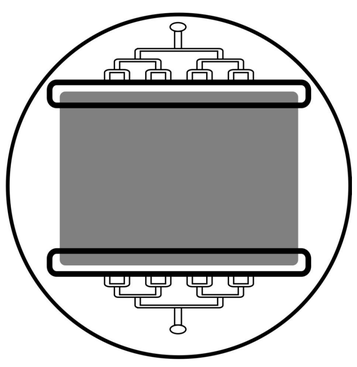
First order reaction rate $r = k(c_{sat} - c)$

Advection-diffusion-reaction $\nabla \cdot (qc) - \nabla \cdot (hD\nabla c) = r$

Erosion $\frac{1-\phi}{\upsilon}\frac{\partial h}{\partial t} = r$

Corresponding experimental set-up



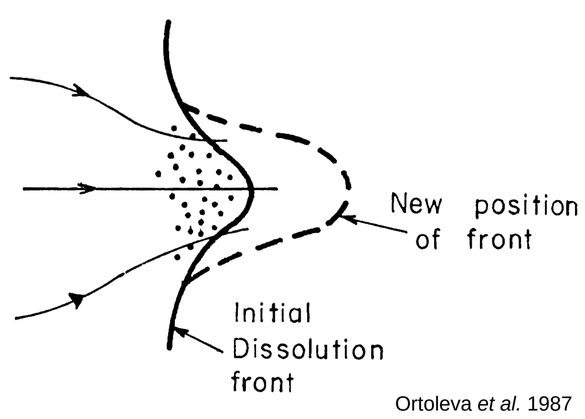


Injection of pure water with a syringe pump

Da and **Pe** set independently by changing the aperture and the flow rate

Flow rate	from 0.25 to 4 mL/hr
Duration	from 1 day to several weeks
Chip size	3cm x 3.5cm

Instabilities of the dissolution front



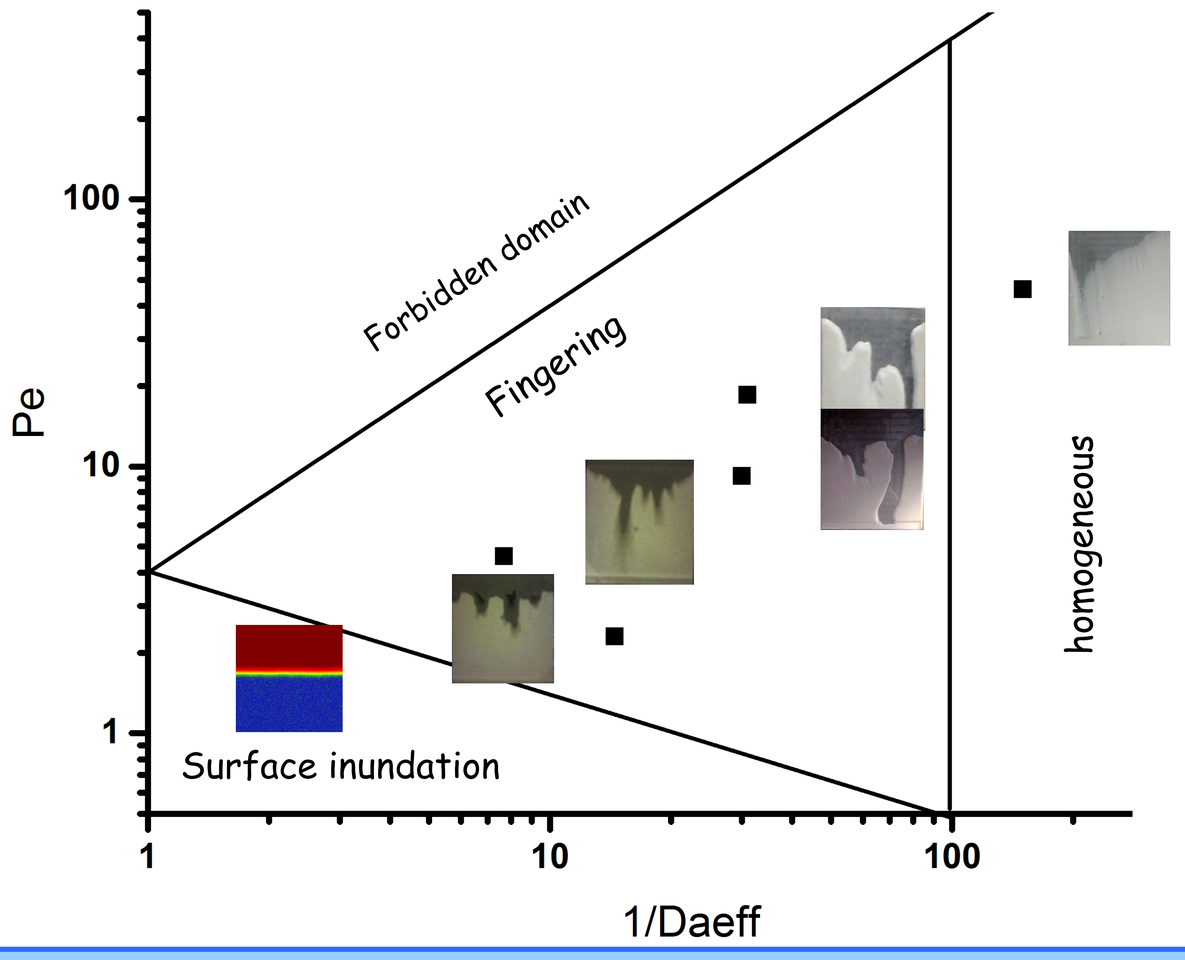
Ortoleva *et al.* 1987

Fundamental numbers

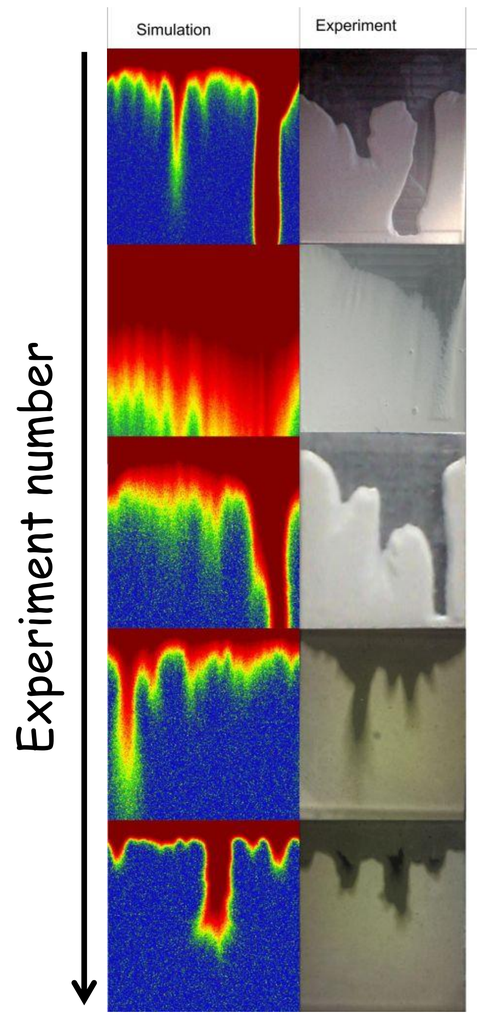
Péclet = $\frac{\text{flow rate}}{\text{diffusion}}$	Controls the thickness of the fingers
Damköhler = $\frac{\text{reaction rate}}{\text{flow rate}}$	Controls the penetration length
$\frac{1}{Da_{eff}} = \frac{1}{Da} + \frac{Pe}{4}$	Effective Damköhler number

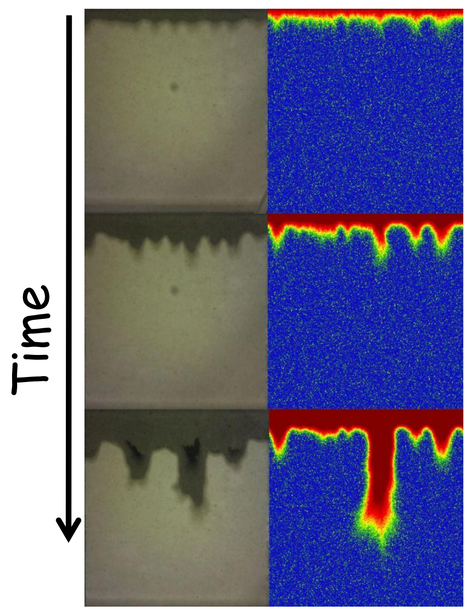
Experimental results and comparison with numerical simulations

Phase diagram as a function of Péclet and effective Damköhler number
- An increase in Pe leads to a thinnning of the fingers
- An increase in Da results in a smaller penetration length
- Domain boundaries are not definitive



Head to head comparison





Numerical simulations

- *Dissol* code from A. Ladd
- Solving by finite elements scheme
- Heterogeneities created with a lognorm distribution

Reproduces exactly the experimental set-up

Good match between experiments and simulations especially on the number of fingers

We have been able to design an experimental set-up allowing to perform completely controlled experiments. All parameters, such as fluid velocity, fracture aperture, roughness, reaction rate… are known. This allows a head-to-head comparison between experiments and numerical simulations bringing very nice results and confidence in the accuracy of the available models and simulations. In the following we need to finish the phase diagram; other developments such as checking the effect of undissolvable obstacles, layered samples, or live interferometry study of the dissolution will be the next step

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