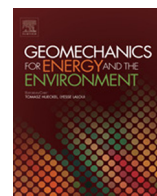




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Hydro-chemo-mechanical coupling in sediments: Localized mineral dissolution

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HIGHLIGHTS

- The paper studies localized dissolution in noncohesive soils under effective stress.
- Sediments become more compressible after localized dissolution.
- The porosity increases within pipes, filled with loose non-soluble quartz.
- Shear wave velocity decreases, and codas indicate increased spatial variability.
- Principal stress rotates in pipes, and sediment reaches Coulomb failure condition.

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ABSTRACT

Mineral dissolution is inherently a chemo-hydro-mechanical coupled process. Field evidence and laboratory results show that dissolution may localize and form open conduits in cohesive media such as carbonate rocks. This study focuses on the evolution of localized dissolution in soils (i.e., frictional and non-cohesive granular materials) under effective confining stresses. Experimental results show the development of localized dissolution (“pipe”) when a carbonate-quartz sand is subjected to reactive fluid flow: only loosely packed quartz grains remain within pipes, and the number of pipes decreases away from the inlet port. Concurrent shear wave velocity measurements show a decrease in stiffness during dissolution due to stress and fabric changes, and more complex signal codas anticipate the development of internal heterogeneity. The discrete element method is used to simulate localized vertical dissolution features in granular materials, under constant vertical stress and zero lateral strain far-field boundaries. As porosity increases along dissolution pipes, vertical load is transferred to the surrounding soils and marked force chains develop. In terms of equivalent stress, principal stress rotation takes place within pipes and the sediment reaches the Coulomb failure condition inside pipes and in the surrounding medium. Dissolution pipes alter the geo-plumbing of the subsurface, enhance fluid transport but limit the long term performance of storage systems, alter the fluid pressure and effective stress fields, soften the sediment and may trigger shear failures.

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1. Introduction

Mineral dissolution and re-precipitation are common processes in the chemical weathering of soils and rocks, and typically involve changes in mineralogy towards more stable compositions within a given geologic environment.

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List of symbols and abbreviations

D	Chemical diffusion coefficient (m^2/s)
D_{10}	Grain diameter (in mm) corresponding to the 10% percentile by weight
D_{50}	Grain diameter (in mm) corresponding to the 50% percentile by weight
Da	Damköhler number
g	Gravitational acceleration
h	Representative vertical length scale
HR	Percentage of rotationally hindered particles in a specimen
i	Hydraulic gradient
ID	Inner diameter
k_0	Coefficient of at-rest lateral earth pressure
k_a	Coefficient of active and passive lateral earth pressure
l	Characteristic length scale of the problem (m)
n	Porosity
Pe	Péclet number
Ri	Richardson number
SF	Fraction of soluble particles
v	Flow velocity (m/s)
V_s	Shear wave velocity
α	Shear wave velocity at $\sigma' = 1$ kPa
β	Stress sensitivity of the shear wave velocity
ε_h	Lateral strain conditions
ϕ	Sediment angle of internal shear strength
θ	Reaction rate constant (1/s)
σ	Effective stress (σ_1 and σ_+ direction of propagation and particle motion; σ_1 and σ_3 minor and major principal stress)

Layering and discontinuities, state of stress, changes in fluid chemistry and gravity-dominated fluid flow contribute to the development of preferential flow paths in sediments and rocks.^{1–3} Vertical tubes are initiated by infiltration, while horizontal tubular cavities result from strata bound water flow.⁴ Dissolution is relatively slow in nature, yet, geological strata that contain carbonates or evaporites are particularly prone to dissolution in high fluid advection systems. Localized dissolution is the cause of sinkholes and settlements in karst regions and calcareous terrains.⁵

Human activity can move natural systems far from equilibrium, such as the injection fluids that change the natural pH and pumping-driven advective transport.^{6,7} Accelerated dissolution rates caused by high hydraulic gradients undermine the performance of dams by increasing uplift pressure and accelerating seepage and piping.^{8–10} The dissociation of load-bearing gas hydrate leads to instability in sediments and has been considered the cause of marine landslides.^{11–14} The geological storage of CO_2 acidifies pore water and causes mineral dissolution that may lead to porosity and mineralogy changes, settlement, and tensile fracturing of the seal rock.^{15–20} Dissolution and precipitation affects permeability changes over time in enhanced geothermal systems.^{21,22} Finally, the consequences of dissolution extend beyond geosystems; for example,

dissolution is one of the main causes of natural deterioration of Portland cement concrete by acid rain,^{23,24} and demineralization makes bones porous and vulnerable to brittle fractures.²⁵

Chemical reaction, advective mass transport, and diffusive transport rates control the evolving topology of dissolution. These can be captured in two dimensionless numbers in terms of flow velocity v (m/s), the characteristic length scale of the problem l (m), the chemical diffusion coefficient D (m^2/s), and the reaction rate constant θ (1/s):

$$\text{Péclet number } Pe = \frac{vl}{D} = \frac{\text{Advective transport rate}}{\text{Diffusive transport rate}} \quad (1)$$

$$\text{Damköhler number } Da = \frac{\theta l}{v} = \frac{\text{Reaction rate}}{\text{Advective transport rate}} \quad (2)$$

Experimental and numerical studies show dissolution topology in terms of Pe and Da numbers:^{26–28} (a) compact dissolution when $Pe < 10^{-3}$, (b) uniform dissolution when $Da < 10^{-3}$, (c) dissolution localization at high $Da > 10^{-2}$ and $Pe > 10^{-2}$ numbers. In addition, density variation in the fluid affects dissolution morphologies due to gravity;²⁹ buoyancy prevails over advection when the Richardson number $Ri = gh/v^2 \geq 1$, where g is the gravitational acceleration and h is a representative vertical length scale.³⁰

Previous studies reveal that homogeneously distributed particle-scale dissolution affects both the small and large-strain properties of sediments:^{31–34} a distinct “honeycomb fabric” emerges,³⁵ vertical displacement and void ratio increase,^{36,33,37–39} the stress ratio between horizontal to vertical effective stress k_0 decreases often to Rankine- k_a failure condition,³⁷ shear wave velocity decreases and attenuation increases,^{33,36} and the peak friction angle decreases.³⁶ Furthermore, fluid permeability changes,⁴⁰ and strain localization may emerge.³⁵ Hu and Hueckel (2007)⁴¹ and Pietruszczak et al. (2006)⁴² introduced important concepts and models of chemo-mechanical couplings for granular contact dissolution and deformation.

This study centers on coupled chemo-hydro-mechanical processes during dissolution in uncemented granular materials under effective confining stress. First, experimental evidence is presented for the emergence of localized dissolution in sediments. Then, discrete element numerical simulations are conducted to gain particle-level insight on the mechanical consequences of localized dissolution in soils.

2. Experimental study

Device and materials. The experimental study is conducted using a quartzitic-carbonate sandy sediment from the Atlantic coastal plains in Georgia, USA ($D_{10} = 0.25$ mm; $D_{50} = 0.8$ mm; 100% passes sieve #10 and is retained on sieve #100. Composition: 52% calcite, 47% quartz, and trace of clay minerals). The chamber is a zero-lateral strain cell (stainless steel cell, ID = 100 mm— Fig. 1); the lower and upper loading plates have a dense groove pattern beneath porous stones to ensure even 1D flow. A linear variable differential transducer LVDT attached to the

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